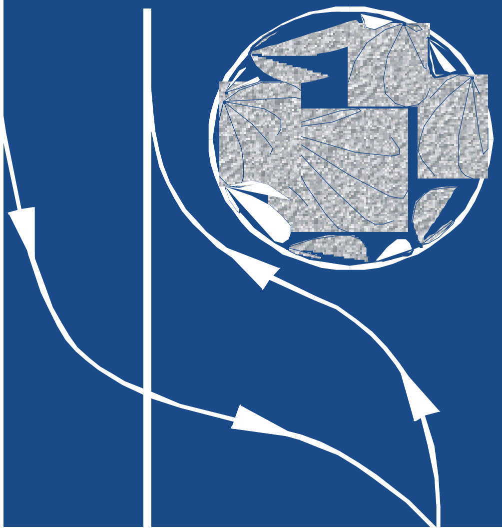

The DOE Complex-Wide Vadose Zone Science & Technology Roadmap

Characterization, Monitoring and Simulation of Subsurface Contaminant Fate and Transport

PRELIMINARY DRAFT

Draft

September 25, 2000

Forward #1

Subsurface hydrology includes the study of water below the land surface, including water held under tension in capillaries and water in saturated zones which freely flows to wells. Subsurface water that lies above the regional water table comprises the vadose zone. Soil physics is an academic discipline that includes the study of water movement in the vadose zone. For the past hundred years or so, soil physicists have been interested in problems of irrigation and drainage of agricultural land. Water below the regional water table is called the phreatic zone, or sometimes simply groundwater. Groundwater hydrology, traditionally has been a discipline of specialists trained in aquifer mechanics, well hydraulics, and groundwater basin management. These specialists were more interested in groundwater supply issues. The two disciplines have evolved separately for the most part, although groundwater hydrologists often learned from the soil physicists and agricultural engineers to solve similar problems, recognizing for example, that the mathematical solution for flow to a drainage canal was similar to that for flow to a stream.

Over the past 20 years, driven in large part by the explosion in regulations to protect groundwater quality, hydrogeologists have faced problems involving flow of chemicals through the vadose zone to aquifers. They looked to the soil science community for tools to compute percolation rates and map migration pathways to the water table. The surge in environmental applications of soil science in the past two decades has led to interdisciplinary research sponsored by a wide range of state and federal agencies. As a result of cross-training and cooperative research, the field of vadose zone hydrology has developed from multiple disciplines including hydrogeology, soil science, civil and environmental engineering, and petroleum engineering, among others. Thus, vadose zone hydrology is quite new; in fact, the first graduate course of this type was offered in the early 1980's.

I was fortunate to be involved with research as the field of vadose zone hydrology was condensing. My experiences as a researcher, which have touched on each of the major areas of research outlined in the vadose zone roadmap herein, illustrate the importance of integrative interactions between site characterization, monitoring, and simulation. With formal training in geology and hydrology, as well as soil physics and civil engineering, in my doctoral program I began to use computer simulations to develop a new tool to determine the saturated hydraulic conductivity of vadose zone soils using borehole infiltration tests. The computer simulations showed surprising results, namely that for deep water table conditions, only a small region surrounding the water filled borehole would saturate no matter how long infiltration occurred. I went to the field in a uniform sand in the semi-arid area north of Socorro, NM to see if this was in fact correct. Using traditional soil physics instrumentation and site characterization techniques I validated the simulations. The simulator proved to be an excellent tool to predict field behavior that never before was recognized.

However, I was surprised to find at this desert site that below about 20 cm depths, the sandy soils were moist, even in the summer. My expectations for dry conditions and my preconceived conceptual model was guided by scientists who thought "there is not direct recharge by rainfall through the vadose zone of semi-arid regions"(Mann; 1976) and who believed that in regions where evaporation plus runoff exceeds precipitation, solutes are retained in the vadose zone (Falconer et al., 1982). In sharp contrast, monitoring of in situ hydraulic head and characterization of hydraulic conductivity at this site revealed that the mean annual recharge rate could be roughly 20% of precipitation.

My graduate students began to establish instrumentation stations throughout the surrounding area and generally found similar results. However, they were surprised that there was quite substantial variability in deep percolation among stations within a single sand dune of essentially uniform composition. The explanation seemed to lie in the importance of lateral flow within the sand dune, as influenced by fine-scale stratification and topography. Field tracer tests confirmed the strong horizontal flow components. However, conventional conceptual models illustrated in the days textbooks on hillslope hydrology show only vertical migration pathways through the vadose zone toward the water table. Using existing sophisticated computer simulators we tried try to reproduce the horizontal flow components, but without success. At about the same time, other graduate students were conducting laboratory experiments designed to test theories on moisture-dependent anisotropy, which were derived from recent stochastic analysis. The success of the laboratory experiments in validating the stochastic theory led us to incorporate this property into the computer simulator. With this new capability, we were able to use the simulator to generally reproduce migration pathways within the sand dune. The site characterization, the laboratory experiments, the theoretical research and the revised computer simulator led us to discover a new process and conceptual model of how water flows in the vadose zone of arguably the most uniform of geologic materials.

Indeed, at DOE sites throughout the country there also have been many surprises of considerably greater importance with respect to flow and transport in vadose zones. Much of what scientists have come to learn about the vadose zone has been achieved only in the last 25 years or so. Undoubtedly, a great deal of discovery of new processes has yet to occur. DOE has the onerous task of ensuring the safety of hazardous and radioactive waste sites through stewardship for perhaps centuries. A discipline integrated vadose zone research program focused on DOE sites over the next 25 years is likely to substantially advance our understanding and ability to more confidently predict subsurface behavior during long term stewardship at DOE facilities.

-Daniel B. Stephens, Chair Vadose Zone Executive Committee; President Daniel B. Stephens & Associates, Inc.; Ph.D. in Hydrology, University of Arizona.

Preface

The project management and logistical aspects of creating a complex-wide vadose zone science and technology roadmap are daunting. Not only did we need to select and assemble a team that could identify the research and development needed over the next quarter century to adequately assess and predict the fate and transport of contaminants in the vadose, but we needed to ensure that the group was well-versed in DOE's needs, the research of other federal agencies, universities and the private sector, and the interests and requirements of federal, state and local governments. This was no small order.

After a lengthy search, 10 internationally recognized individuals were selected, and accepted, positions on the Vadose Zone Roadmap Executive Committee, with the charter to develop a preliminary draft roadmap by September 25, 2000. Four of these individuals also accepted the challenge to serve as workgroup chairs and to assemble the remainder of the participants. The final team consists of 62 individuals, many internationally recognized, vadose zone experts—and authors of most of the vadose zone books on the market. These individuals (a complete list is provided in Appendix A) represent a cross section of DOE national laboratories, other federal agencies, industry, academia, and the international community.

As a starting point for this group, INEEL and other DOE national laboratory personnel familiar with end user needs and vadose zone issues (including several key collaborators developing the *Vadose Zone Science and Technology Solutions* book) prepared a summary of the vadose zone characterization, monitoring and modeling deficiencies and capability gaps across the complex. The results of this effort, entitled D*raft Vadose Zone Science Integration and Technical Basis*, was provided to all roadmapping participants.

On March 30, 2000, the Vadose Zone Roadmap executive committee assembled for the first time to begin its task. Over the next six months, this committee and four (ultimately condensed to three) roadmap workgroups formed the multi-disciplinary team that has outlined in these pages the vadose zone research priorities for the next quarter century.

This is a map drawn in broad strokes, leaving many roads still unmarked. Nevertheless, it provides the first guidance at a federal level for the path forward through this heretofore-unmapped land. If successful, the research described in this document will lead dramatically improved characterization, monitoring and modeling capabilities that will provide DOE with the scientific foundation upon which regulators and site managers can make decisions and predictions about the vadose zone with great confidence.

Additional work to integrate and sequence the research priorities identified in this roadmap will require continued dialogue within the vadose zone research community, with other U.S. government and state and local agencies, and with affected stakeholders. In particular, further effort is needed to broaden consensus within the subsurface science research community and site managers on the goals identified here, to integrate the necessary research around issues that cut across scientific disciplines, and to discuss the implications that a vastly improved predictive capability will have on policy makers, regulators, and the U.S. public.

- Stephen J. Kowall, Idaho National Engineering and Environmental Laboratory

TABLE OF CONTENTS

EXECUTIVE SUMMARY

The vadose zone comprises the region lying between the earth's surface and the top of the groundwater. Over the last several decades, the U.S. Department of Energy (DOE) has conducted energy research and weapons development and production, which has led to the introduction of toxic chemicals into the vadose zone. Until recently, contamination in the vadose zone was believed to remain relatively immobile. Thus, little attention was paid to understanding the nature of the vadose zone, or the potential pathways for contaminants to migrate through the zone. However, recent discoveries of toxic chemicals migrating great distances through the vadose have changed many assumptions, both about the vadose zone and the importance we place on understanding this region.

As a result, DOE tasked the Idaho National Engineering and Environmental Laboratory (INEEL) with development of a science and technology roadmap focused on identifying research spanning the next 25 years that would be necessary to be able to better predict the fate and transport of contaminants in the vadose zone. This in turn will provide the basis for reducing scientific uncertainty in environmental remediation, waste handling and long-term stewardship decisions across the DOE complex.

Science and technology roadmapping is a form of strategic technology planning used by an increasing number of companies, industries, and U.S. government agencies. The purpose of science and technology roadmaps is to develop a common perspective on possible future science and technology needs, in an effort to help make better research and development (R&D) investment decisions.

Between March 30 and September 15, 2000 an executive committee and four roadmapping workgroups (eventually consolidated into three workgroups) comprised of sixty-two representatives from DOE, National Laboratories, other U.S. Government agencies, universities and industry[1], discussed in detail their vision for a set of science and technology capabilities that would transform the scientific basis for making decisions involving the vadose zone. The resulting roadmap, provides a structure for understanding the context of remedial and long-term stewardship decision-making by DOE, the components that contribute to an improved scientific input to these decisions with respect to the vadose zone, and a discussion of the critical research that must be conducted over the next 25 years necessary to realize the vision of better decision making. If successful, the research described in this document will support DOE's complex-wide needs while simultaneously advancing the state-of-the-art of characterization, monitoring and modeling science and technology.

The need for a vadose zone roadmap is driven by the following:

- **Unpleasant Surprises.** Recent evidence indicates that hazardous and radioactive chemicals have migrated unexpectedly through the vadose zone.

- **Early Intervention is Best.** The ability to identify and predict contaminant pathways in

[1] A complete list of participants is provided in Appendix A.

the vadose zone, before they have a chance to migrate to the water table is exponentially more cost effective, and offers similarly large potential to minimize risks to human and environmental health.

- **Existing Capabilities are Insufficient.** At this time there is no single predictive tool that has been demonstrated to reliably simulate fate and transport of organic and inorganic chemicals and radionuclides in the vadose zone. (need something about sensors and instrumentation here also) Without one, DOE cannot make rational and publicly acceptable decisions regarding short-term remediation or long-term stewardship.

- **Investment and Integration is Necessary.** The inherently complex nature of the vadose zone demands an integrated and multidisciplinary research approach, which will require increased investment and a long-term focus.

Creating a better scientific foundation for decision-making, with respect to the vadose zone, will require increased knowledge and capabilities in three areas: (1) understanding basic subsurface processes; (2) better data collection and monitoring capabilities; and (3) new computer models and predictive capabilities. The specific research required over the next 25 years in each of these areas forms the centerpiece of the roadmap. It is organized to emphasize key knowledge and capability gaps including: physical descriptions of flow and transport, chemical properties and processes, biological properties and processes, colloidal properties and processes, multi-phase, chaotic and unstable processes, sensors and instrumentation, network design and systems optimization, computer hardware, software and algorithms, and finally, integration issues including scaling, uncertainty and the value of data.

Over the next few decades, dramatic and fundamental advances in computing, communication, electronics and microengineered systems will transform many of the scientific and technical challenges we face today. It is not unrealistic to imagine that by the year 2025 vadose zone researchers will have microscopic sensors on devices the size of rice grains that can be injected into the earth, and that are so sensitive that they will be able to determine with certainty whether or not contaminant migration is occurring on a time-scale that today would take centuries to recognize. Finally, that we will have simulation capabilities comparable to those currently used by DOE to model nuclear weapons systems and molecular processes in biology, thus providing an understanding of the vadose zone that is sufficient to make scientifically sound public policy and regulatory decisions.

In addition to identifying areas of priority research, this roadmap addresses a number of key changes in supporting infrastructure and research approach that DOE must adopt:

- **Integrating Research and Integrated Field Experiments.** Integration of current and future research will be critical to understanding the basic processes at work in the vadose zone. Integrated field experiments, organized to address vadose zone questions that are most important to DOE's ability to assess and predict contaminant fate and transport, offer a means to foster such integration and dramatically accelerate advances in the application of vadose zone research.

- **A "Green Machine."** Visualizing vadose zone characteristics and modeling its behaviors is a problem of comparable difficulty and complexity as that of simulating the explosions and operations of nuclear weapons and molecular processes in biology. Thus there is a need for access to a massively parallel computing capability, or "Green Machine," that would revolutionize current abilities to assess and predict contaminant fate and transport in the vadose zone.

- **Data and Model Libraries.** Widespread distribution and use of vadose zone experimental results and models suffers from the lack of better methods for collecting and disseminating existing information and models. "Libraries" of such information (whether virtual or real) that would place the results of current experiments in context, and in the hands of those who might benefit from them, are greatly desired by the vadose zone research community.

- **National Leadership: A Vadose Zone "Czar."** The complexity of the scientific challenge of adequately monitoring and predicting vadose zone properties and processes, coupled with the number and variety of stakeholders who care about these issues, drives the need for national leadership to create and guide a multi-agency, multi-year, long-term and integrated approach to the vadose zone challenge.

1.0 INTRODUCTION

The vadose zone is the region lying between the earth's surface and the top of the groundwater table. Until recently, contamination in the vadose zone across the U.S. Department of Energy (DOE) complex (the residue of many years of energy research and weapons development) was believed to remain relatively immobile within the vadose zone. Thus, little attention was paid to understanding the nature of the vadose zone, the potential pathways for fluids or contaminants to disperse throughout the zone, or the myriad possible interactions among fluids, contaminants, and the chemical, biological and physical components of the region. However, new discoveries of hazardous and radioactive chemicals migrating through the vadose zone and entering underlying aquifers have changed many assumptions, both about the vadose zone and the importance we place on understanding this region.

As a result, the DOE directed the Idaho National Engineering and Environmental Laboratory (INEEL) to develop a science and technology roadmap to identify the research needed over the next 25 years. The roadmap considers the research that will improve predictions of fate and transport of contaminants in the vadose zone, and thus reduce uncertainty in decision-making across the DOE complex. Science and technology roadmapping is a form of strategic technology planning used by an increasing number of companies, industries, and U.S. government agencies. The purpose of science and technology roadmaps is to develop a common perspective on possible future science and technology needs, in an effort to help make better research and development (R&D) investment decisions. Science and technology roadmaps serve as pathways to the future. They call attention to future needs for development in underpinning science and applied technology, provide a structure for organizing technology forecasts and programs, and communicate scientific technological needs and expectations among end-users and the R&D community.

On March 30, 2000, an executive committee formed to address the science and technology for characterizing, monitoring, and modeling subsurface contaminant fate and transport. Over the next six months, this committee and four roadmapping workgroups comprised of sixty-two representatives from DOE, National Laboratories, other U.S. Government agencies, universities and industry[2], discussed in detail their vision for a set of science and technology capabilities that would transform the scientific basis for making decisions involving the vadose zone. The resulting roadmap, presented in the following pages, provides a structure for understanding the context of remedial and long-term stewardship decision-making by DOE, the components that contribute to an improved scientific input to these decisions with respect to the vadose zone, and a discussion of the critical research that must be conducted over the next 25 years to realize the vision of a sound scientific basis for making public policy and regulatory decisions. If successful, the research described in this document will support DOE's complex-wide needs while simultaneously advancing the state-of-the-art of vadose zone characterization, monitoring and modeling science and technology.

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[2] A complete list of participants is provided in Appendix A.

1.1 The Need for a Complex-Wide Vadose Zone S&T Roadmap

The DOE has conducted energy research and weapons development and production at facilities in 31 states and in Puerto Rico. Toxic chemicals generated at these sites have been introduced into the underlying soils and aquifers as a result of a number of historical and current practices. At present, more than six billion cubic meters of subsurface media at 134 sites are contaminated[3]. Of this approximately 700 million cubic meters is groundwater contaminated with organic wastes (e.g., solvents, fuels, PCBs), radioactive waste, or mixed (chemical and radioactive) radioactive waste. Typically, the groundwater contamination results from the migration of contaminants through the vadose zone. Today, some 60 million cubic meters of soil and rock comprising the vadose zone are contaminated, mostly with radioactive waste or mixed low-level radioactive waste.[4]

Unpleasant Surprises. Anticipating the failure of some aspect of a long-term stewardship solution is not unreasonable: recent evidence indicates that hazardous and radioactive chemicals have migrated unexpectedly through the vadose zone at multiple DOE facilities. For example, at the Hanford 200 Area tank farm, technetium-99 migrated to ground water through 200 to 300 feet of what was previously believed to be highly sorptive material that would have prevented such migration. At Los Alamos National Laboratory, plutonium and americium were discovered 100 feet beneath a liquid waste impoundment where nuclide transport was believed to be dominated by sorption and thus radionuclide migration should have been very limited. At the Sandia National Laboratory TCE from a landfill have been discovered at depths of 500 feet in an area of very dry soil and low recharge. At the high-level radioactive waste disposal repository in the dry desert climate of Yucca Mountain, modern tracers, bomb pulse chlorine-36 and tritium, were found in fractured tuff at depths of as much as 1200 feet, suggesting rapid recharge along preferential pathways.

Information of this type led the NRC to review DOE's long-term stewardship program and find gaps in basic scientific understanding, including "deficiencies in our ability to make accurate estimates of subsurface contaminant behavior, especially in the conceptual understanding of this behavior to enable accurate and robust modeling". This conclusion echoes that of the GAO in their report on Hanford concerning DOE's inability to predict fate and transport in the subsurface with the certainty required to make key environmental decisions.

Early Intervention is Best. In almost all circumstances the most effective way to prevent potentially dangerous situations from escalating into crises is to intervene early. This is true with respect to oil or other hazardous materials spills for which rapid containment and cleanup is the law. It is also true in the case of many diseases for which prevention, early diagnosis, and early treatment are often critical to survival. It is especially true for all types of environmental contamination, where early remediation measurably reduces risks to workers, the public, and the environment.

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[3] *Research Needs in Subsurface Science*, published by the National Research Council in 2000, pp.15 & 21

[4] From Cleanup to Stewardship, U.S. Department of Energy, 1999, p. X.

The value of early intervention can be roughly quantified. Extensive field experience and theoretical considerations of a typical landfill or cocooned building, for example, suggest that remediation costs and risks are reduced with smaller volumes, greater concentrations, and when intervention occurs when waste materials have only migrated into the vadose zone but not yet reached the saturated zone. The relationships among these factors is familiar from the theory of entropy and from experience with enrichment of uranium. Further more, these relationships are exponential rather than linear—making remediation of pollutants at their source by far the least expensive and least risky approach to environmental and human health protection. The next best alternative, from a risk, cost and difficulty standpoint, is stabilization and removal while the contaminants are in the vadose zone, before the saturated zone (groundwater) has been affected. Understanding how contaminants are transported within the vadose zone; having the ability to locate, characterize and quantify them; and ultimately being able to remove or stabilize pollutants before they leave the vadose zone is a less-risky, lower-cost strategy than one that waits until pollutants have reached the saturated zone.

Existing Capabilities are Insufficient. The surprises described earlier, and many others, are a clear indication that our current understanding and capabilities related to the vadose zone are inadequate. Our ability to characterize the apatial distribution of chemicals and migration pathways is quite limited. The state-of-the-practice in vadose zone monitoring to detect chemical migration is limited primarily to in situ samplers which retrieve pore-water only from limited volumes of relatively wet soils, and to geophysical methods which only indirectly measure waste movement. There is no technology available to ensure detection of migration via discrete, narrow fingers in unsaturated porous media, or to sample fluids coating only portions of fracture surfaces, or to collect colloidal particles from the pore water in the vadose zone.

In addition, field investigations often reveal that the extent of chemical migration is poorly predicted with mathematical models of flow and transport in the vadose zone. This poor record may be attributed to incomplete understanding or unrecognized physical/chemical/ biological processes, insufficient or inaccurate characterization of the processes or site properties, and inadequate or inaccurate numerical models. At present, there is no single predictive tool that has been demonstrated to reliably simulate fate and transport of organic and inorganic chemicals and radionuclides in thick vadose zones comprised of porous and fractured media under variably saturated conditions. Without these capabilities, DOE cannot make rational and publicly acceptable decisions regarding short-term remediation or long-term stewardship. In their most recent report, the NRC concurs, stating that, “mathematical modeling of contaminant fate and transport is an essential tool for long-term institutional management, but its track record to date at DOE sites, particularly where contaminants reside in the unsaturated, or ‘vadose zone’, has been mixed.”[5]

Uncertainty Is Creating Public Distrust. The inability to explain basic subsurface processes and predict contaminant fate and transport in the vadose zone have led regulators to fall back on the most conservative expectations: all contaminants will move rapidly through the vadose zone to the groundwater. In the late 1960s travel time of C-14 from the surface to ground

[5] formal citation to come

water at the INEEL was estimated at 80,000 years; today the estimates are closer to 50 years.[6] Similar foreshortening of travel time predictions for other contaminants has also occurred. Despite these dramatic changes in expectations, many in the scientific/technical community assert that we are no more certain of our current predictions than we were of those made three decades ago.

The scientific uncertainty that characterizes our understanding of the vadose zone has two additional consequences: erosion of the public trust and greatly increased public expense. For example, at some sites with thick, low permeability vadose zones, DOE is not allowed to take credit for any concentration attenuation that might naturally occur. Instead regulators require extremely conservative risk assessments by assuming all chemicals will instantaneously reach the aquifer below, completely ignoring any mitigating effects of dispersion and sorption through the vadose zone. Consequently, engineered facilities arguably are being over-designed, sometimes at excessive expense. At sites where the remedy or containment plan ignores vadose zone processes, monitoring programs rely exclusively on the deep groundwater, which, if found to be affected would require expensive remediation efforts that could have been avoided through vadose zone monitoring and remediation. Through this vadose zone science and technology roadmap we will build a technical foundation to give regulators and the public confidence that vadose zone characterization, modeling and monitoring can support DOE decisions on remediation and stewardship.

Investment and Integration is Necessary. While investigations of saturated groundwater flow and transport have received substantial attention in the form of R&D funding from DOE and other agencies, vadose zone research has been much less plentiful. Research that does exist is fragmented and scattered among multiple disciplines. In part, this reflects the inherently more complex nature of the vadose zone—a function of the extreme heterogeneity of hydraulic properties at variable moisture conditions, the multiple fluid phases where chemical partitioning and biodegradation occur, the highly non-linear and strongly coupled systems, the transient surface boundary conditions, and the unstable nature of flow phenomena, among others. The comparatively small emphasis placed on vadose zone research is also a function of the need for integration and multidisciplinary approaches to this research—vadose zone research problems are not effectively dealt with as isolated \$100,000 to \$200,000 projects typical of DOE research efforts. The complexities of the vadose zone increasingly demand the integration of many scientific disciplines in a coherent fashion. Yet, as will be discussed further in Section 3.0, there is at present no such focused integrated research program within the United States.

Finally, vadose zone research has suffered with the rest of DOE's Environmental Management (EM) R&D budget, which has been declining over the past three years. This despite the fact that, as discussed above, improved scientific understanding and monitoring of the vadose zone holds the greatest potential for reducing costs and future human health and environmental risks across the country.

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[6] For a further discussion of predicted contaminant travel times and how they have changed, see *Research Needs in Subsurface Science*, published by the National Research Council in 2000, p.30.

1.2 A Roadmap for Reducing Uncertainty

Decisions regarding remediation strategies and long-term stewardship across the DOE complex involve many interests:

- external stakeholders, regulators, science and technology participants, and the public, whose decision-making is driven by perceived risks to human health and the environment;
- the DOE site management community and science and technology users, who make decisions regarding remediation, waste handling, decommissioning, remedial monitoring, and stewardship in response to regulation, available technology, cost and time constraints; and
- the science and technology community and science and technology providers, whose knowledge and predictive capabilities should provide the technical foundation for the decisions of the other two groups.

Creating a better scientific foundation for making both near-term remediation and waste handling decisions and long-term stewardship decisions regarding the management of contaminated DOE sites with respect to the vadose zone will require:

1) an enhanced understanding of basic subsurface process;
2) adequate data on the extent and character of existing contamination, and the ability to monitor it effectively and;
3) the ability to translate this understanding and data into new predictive models that can minimize the technical uncertainty in environmental management decisions.

Each of these areas can be further subdivided. Thus, to develop an enhanced understanding of basic subsurface processes, a roadmapping team was assembled that subsequently identified six key areas for which DOE needs improved capabilities: physical description of flow and transport processes; chemical properties and processes; biological properties and processes; colloidal processes; multiphase flow and transport processes; and chaotic and unstable processes.

Similarly, acquiring the necessary characterization and monitoring data was subdivided by the workgroups into the following seven capabilities: the geological, chemical, biological, hydrological (GCBH) framework; chemical characterization; biological characterization; hydrological properties and processes; network design and system optimization; sensors & instrumentation; and integration issues. Finally, seven key components of modeling and simulation were identified: software, hardware, numerics, uncertainty, scaling, strongly coupled systems, and integration and validation.

This organization, depicted in Figure 1 illustrates the "highways" of this Roadmap. For each area, the Roadmap identifies important paths that will lead to the tools and fundamental scientific understanding of the vadose zone that stakeholders and the DOE site management community need to make sound decisions. Interim destinations along this roadmap are discussed in Section

2.0 of this roadmap in the context of three timeframes: what must be done now to reach goals identified for the year 2004, the R&D that must be accomplished to meet goals established for 2010, and the longer-term research that must occur to meet goals for the year 2025 and beyond. Details on the sequencing and integration of this research is provided in Appendices C and D.

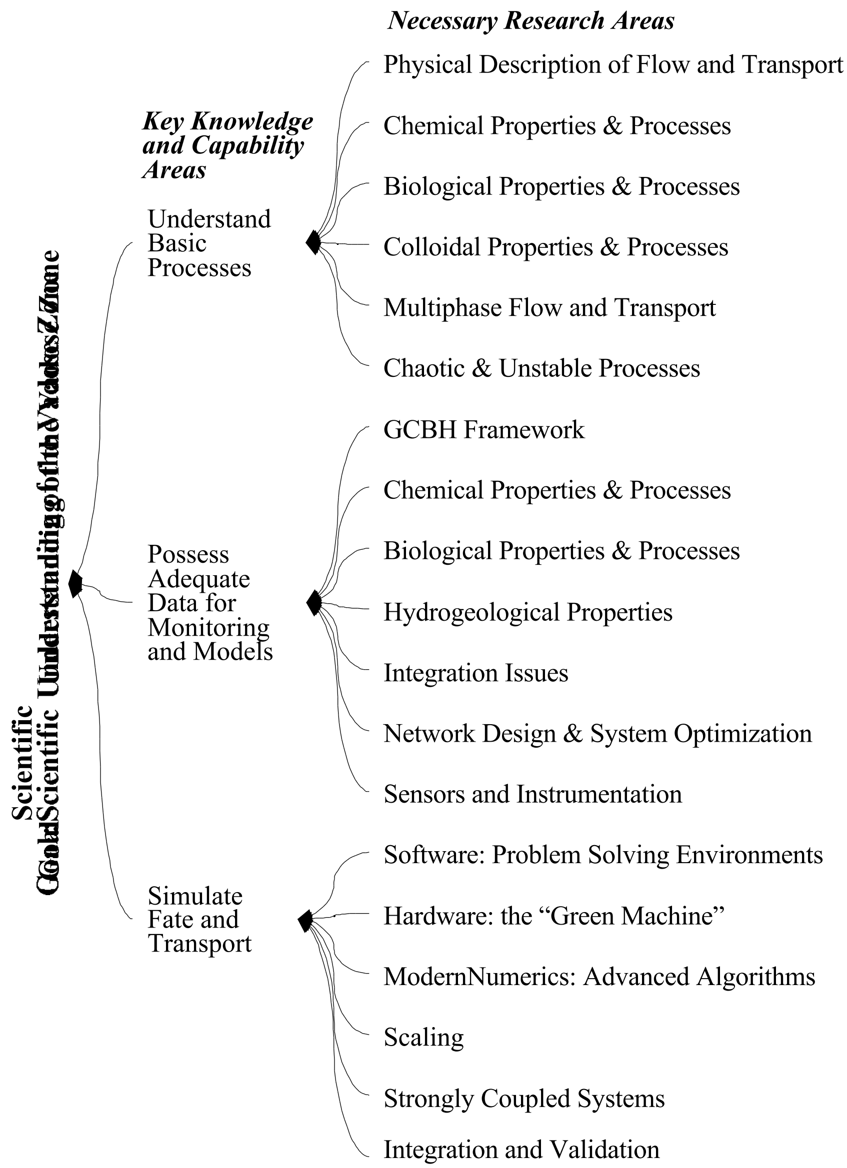

Figure 1. The Roadmap Highway

1.3 A (Grand) Vision for 2025

Over the next few decades, dramatic and fundamental advances in computing, communication, electronics and microengineered systems will transform many of the scientific and technical challenges we face today. For example, sensors that once would have required rooms of complex and fragile equipment are now located on a microchip. The computing power once reserved to put a man on the moon, is now far surpassed by the power found in a personal home computer. Moore's Law, which has predicted the doubling of computing power every 18 months, forecasts the availability of more than a thousand fold increase in computing speed within the next 15 years, and a more than a hundred-thousand fold increase by the year 2025.

With these types of advances becoming commonplace, one can envision by the year 2025:

> **A new generation of microscopic sensors,** the size of a grain of rice, that minimize the sensors' impacts on the measurement, that will increase the density of measurement while lowering the cost of individual measurements, that can make multi-channel measurements (e.g., chemical species detection or pressure, temperature, pH or eH measurements) simultaneously in real time. These sensors may be eventually small enough to inject into the material being investigated;
>
> **Ultra-sensitive monitoring capabilities** that allow investigators to determine with certainty whether or not contaminant migration is occurring on a time-scale that today would take centuries to recognize.
>
> **Vadose zone simulation abilities** comparable in their complexity, speed, and accuracy to those used by DOE to model nuclear weapons systems and explosions or molecular processes in biology.
>
> **A fundamental understanding of vadose zone properties and processes** that is sufficient and robust enough so that scientific uncertainty is no longer a large component of the public and regulatory debate.

If the items highlighted above describe some of the capabilities awaiting us in the next quarter century, those below address some of the key changes in approach DOE must adopt to make the vision a reality.

Integrating Research and Integrated Field Experiments. Integration of current and future research will be critical to understanding the basic processes at work in the vadose zone, for capturing the data necessary for monitoring contaminant migration and feeding computer simulations, and developing models capable of predicting contaminant behavior and fluid flows. Integrated field experiments, organized to address vadose zone questions that are most important to DOE's ability to assess and predict contaminant fate and transport, offer a means to foster such integration and dramatically accelerate application of vadose zone research results.

A "Green Machine." Visualizing vadose zone characteristics and modeling its behaviors is as difficult and complex as simulating nuclear weapons tests and molecular processes in biology.

A massively parallel computing capability, or "Green Machine," is needed to provide computing power capable of revolutionizing our current ability to predict contaminant fate and transport in the vadose zone.

Data and Model Libraries. Widespread distribution and use of vadose zone experimental results and models research is needed, to put the results of experiments and research in the hands of those who might benefit from them as evidenced by the recent publication of Vadose Zone Science and Technology Solutions.[7].

National Leadership: A Vadose Zone "Czar." The complexity of the scientific challenge of adequately monitoring and predicting vadose zone properties and processes, coupled with the number and variety of stakeholders who care about these issues, drives the need for national leadership to create and guide a multi-agency, multi-year, long-term and integrated approach to the vadose zone challenge.

Section 2.0 of this Roadmap outlines vadose zone research priorities between now and 2025 needed to make this grand vision a reality. Section 3.0 suggests complementary changes in research approach and infrastructure.

[7] Formal citation to come

2.0 TOWARD A BETTER SCIENTIFIC UNDERSTANDING OF THE VADOSE ZONE

As outlined in the Introduction to this roadmap, significant advances must be made in three areas to provide decision-makers with the information needed to make good remediation, monitoring, post monitoring, and stewardship decisions. Each of these areas: *Understanding Basic Process*; *Possessing Adequate Data and Monitoring Capabilities; and, Developing Adequate Models and Simulation Capabilities*, is discussed in detail below. The tables, which follow each portion of this section, represent the "raw input" provided by workgroup members during the development of the roadmap.

2.1 Understanding Basic Processes

The following pages outline the knowledge gaps in physical, chemical, and microbiological processes that are currently a major source of uncertainty when addressing remediation and long-term stewardship technologies. Advances in our conceptual understanding of the basic fluid flow and contaminant transport and transformation processes are closely connected with, and cannot be considered separately from, advances in numerical modeling, computer hardware, and the characterization and measurement of subsurface properties and processes. This integration of processes with numerical modeling and characterization is also important for prioritizing which gaps in process understanding must be addressed in both short-term and long-term research. Integration is also essential to identifying which properties are the most relevant, and at what resolution, accuracy, and uncertainty they must be estimated to be effective inputs to predictive models.

There are six important categories of basic process research: (1) the physical description of flow and transport processes, (2) chemical processes, (3) microbial processes, (4) colloidal processes, (5) multiphase flow and transport processes, and (6) chaotic and unstable processes. This separation along traditional disciplinary lines is purely arbitrary, and was implemented only for organizational purposes. In reality, fluid flow, contaminant transport, and biogeochemical processes do not occur in isolation but take place in a nonlinear and tightly coupled manner. For example, while fluid flow is the primary mode of transport for most contaminants, the chemical makeup (concentration and ionic composition) of the fluid phase and various biogeochemical reactions can significantly affect fluid and solid phase properties, which in turn affects fluid velocities and rates of contaminant transport. Improved descriptions of the nonlinear transient interactions between the various processes and subsurface properties are needed as well as methods to deal with the overwhelming complexity of these processes.

2.1.1 Physical Description of Flow and Transport Processes

The scientific community generally recognizes that our ability to predict and optimally manage fluid flow and contaminant transport processes in the vadose zone is limited by inadequate understanding of many fundamental liquid-gas-solid media interactions, as well as by a general lack of methodologies to represent flow processes across different spatial and temporal scales. The latter is an immediate consequence of the overwhelming heterogeneity of the hydraulic and transport properties of the subsurface. Flow and transport processes that are well captured by macroscopic continuum approaches at relatively large spatial scales in homogeneous media may be critically misrepresented at smaller scales in highly heterogeneous granular or fractured porous media. While our knowledge of vadose zone flow and transport processes has advanced enormously over the past several decades, current knowledge remains largely inadequate to address all of DOE's subsurface remediation and long-term stewardship responsibilities. In the near-term it is essential to narrow the gap between the state-of-the-art and the state-of-the-practice. For this reason, by 2004, known models and solutions based on a better understanding of physical and biogeochemical processes will need to be incorporated in DOE flow and transport prediction models and decision making.

There are many obstacles to obtaining and implementing more realistic process-based descriptions of fluid flow and solute transport. Improved descriptions are needed of many pore-scale processes and associated liquid-solid and liquid-gas interactions. Driving forces affecting flow and transport (especially chemical gradients) need to be studied. More general constitutive relationships between fluid, geochemical and solid phase parameters and variables, including two-phase air-water hydraulic property representations of unsaturated media under extremely dry conditions also require attention.

Improved process-based descriptions of flow and transport in macroporous soils and unsaturated fractured media represent an especially significant challenge. What is needed are more realistic representations of pore-space geometry of such structured media using emerging geologic, pedologic, and topologic characterization techniques. In parallel with this effort, improved descriptions of matrix-fracture interactions of structured media must be developed.

The propagation of physical processes across scales and the associated prediction uncertainty affects all modeling activities in earth systems. Not only is it necessary to match the proper model to the scale of interest, there is also a need to understand how to move from one scale to another without losing key elements of the basic processes involved. This entails different definitions and characterization methods for transport properties, and different ways for representation and propagation of driving forces. A related challenge is to develop a formalized methodology that permits significant data at different scales to be both recognized and incorporated in the formulation of a robust conceptual model of key flow and transport processes operating at a scale relevant to DOE's remediation, containment, and long-term stewardship responsibilities. This will require methods for efficient assimilation of different types of data having different spatial and temporal resolutions, different geostatistical properties, and different measurement errors.

It is important to recognize the need for proper data at a range of scales. Applying models to real-world situations will require hypotheses about the main processes operative at individual sites and estimates of site-specific properties. These processes and parameters include (1) hydraulic and solute transport parameters (such as the capillary-pressure saturation curves, liquid and gas conductivities as a function of phase saturation, and various diffusion coefficients, tortuosities, and dispersivities), (2) fluid properties (such as density, viscosity, enthalpy, vapor pressure, surface tension, and wetting angle), (3) thermal properties (such as heat capacities and the saturation dependent bulk thermal conductivity), (4) biogeochemical parameters (such as thermodynamic and kinetic data for homogeneous and heterogeneous reactions involving aqueous and gaseous species and minerals, and mineral surface areas), (5) initial and boundary conditions (for fluxes, temperature, pressures, fluid compositions, aqueous and gas phase compositions, and mineral concentrations), and (6) fluid and chemical sources and sinks.

Research Priorities: Physical Description of Flow and Transport Processes	
2004	▪ Review the state-of-the-practice in DOE for site characterization and remediation, and integrate known models/modules with a sounder physical-chemical basis (state-of-the-science) into DOE flow and transport prediction models and solutions. ▪ Identify and assess the importance of fluid flow and chemical transport processes and subsurface properties relevant to site characterization, remediation and long-term stewardship (e.g. gravitational, pressure, temperature and chemical gradients). ▪ Identify and prioritize critical gaps in the understanding of vadose zone flow and transport processes relevant to the DOE environmental restoration and long-term stewardship missions. This would form a basis for the design and implementation of subsequent laboratory and field experiments to resolve these gaps by 2010. ▪ Develop methods to quantify spatial variability in flow and transport parameters, fluid properties, thermal properties, biological and chemical reactions. ▪ Develop and implement improved models for evapotranspiration and root water uptake as a function of water stress and other factors. ▪ Develop process-hierarchical approaches for describing and modeling controlling processes (e.g., models of increased complexity for flow in structured media, or increasingly sophisticated process-based approximations for gaseous transport or the flow of non-aqueous phase liquids).
2010	▪ Design, implement and analyze controlled laboratory and field experiments to develop the key databases and understanding needed to resolve knowledge gaps in flow and transport processes operative in granular media. ▪ Develop realistic representations of pore-space geometry of fractured media using geologic, pedologic, and topologic characterization techniques. In parallel, develop and implement improved descriptions of matrix-fracture interactions. ▪ Develop general constitutive relationships for both granular and structured media. ▪ Develop methods for characterizing and propagating physical processes and properties, and their uncertainties, across different scales (governing equations, driving forces, and/or media properties). ▪ Develop methods for assimilating data from different scales; identify linkages between relevant small-scale processes and properties with larger-scale flow and transport behavior for cases where no simple upscaling is possible. ▪ Integrate critical elements of chemistry and microbial activity in models for liquid flow and solute transport. ▪ Develop pore network models based upon appropriate equations (e.g. Navier-Stokes), and devise methods for upscaling processes and/or properties to sample and formation scales.
2025	▪ Develop process- and scale-adaptive models for modeling unsaturated flow and

	transport in different types of porous media. Models should invoke processes at the appropriate scale and employ the corresponding transport and hydraulic properties. ▪ Incorporate solid surface heterogeneity (pore and sample scale) with detailed hydrodynamics into solute transport models (to model opportunity times for reactions, streaming potentials, and other processes or variables). ▪ Incorporate microbiology modules into physical models of solid-gas-liquid environments. ▪ Incorporate solid surface evolution due to biogeochemical reactions (precipitation-dissolution, reduction-oxidation, and other reactions that can change solid-phase geometry and density of exchange sites) into transport models necessary for long time-scale simulations.

2.1.2 Chemical Properties and Processes

Numerical continuum models of reactive flow and transport in heterogeneous, variably saturated, nonisothermal porous media have advanced considerably in the past several years. It is now practical to carry out simulations in one- and two-spatial dimensions for coupled, transient, multiphase (aqueous, gaseous, CO_2, and NAPL), multicomponent (on the order of 10-20 degrees of freedom) flow and transport on small to medium sized grids (2,500-10,000 nodes). Processes that can now be incorporated in such calculations include: homogeneous reactions consisting of aqueous complexing, ion pairing, and redox reactions; heterogeneous reactions consisting of mineral precipitation and dissolution, sorption, and aqueous-gaseous reactions; and transport and reaction with colloids. Reaction rates may be described either through kinetic rate laws or local equilibrium, as appropriate. In addition, flow and transport may be sequentially coupled, thus accounting for changes in porosity and permeability resulting from chemical reactions. Few, if any computer codes currently account for all of these different features. While limited work on developing next-generation codes for three-dimensional simulations with large numbers of unknowns based on massively parallel computing architectures has begun, optimal development of parallel algorithms is still in its infancy.

First, chemical parameters that determine the characteristics of contaminated solutions, and the rates at which they will react with local site characteristics, include thermodynamic properties of solute species. Algorithms and databases for modeling aqueous solution chemistry exist at 25 C and ionic strengths less than approximately 1M using the extended Debye-Huckel activity coefficient correction. At higher temperatures and high ionic strengths, thermodynamic data becomes more sparse. While obtaining data for these properties requires very stringent laboratory conditions, they can be readily obtained with existing technologies.

Second, rate laws are commonly based on transition state theory or simple reaction kinetics. Rate constants for kinetic descriptions of mineral reactions have been obtained over a limited temperature range and for limited compositional variables, primarily pH. Often, however, rate laws measured in the laboratory are not suitable for direct use in a reactive transport model because they do not cover a suitable range of conditions. For example, the oxidation state of the system may change from oxidizing to reducing along the flow path. A single rate law is needed which encompasses the variation in redox and other conditions over the entire flow path.

Furthermore, mineral rate laws apply to crystalline phases and not glasses or phases with mixed structural states such as mixed layer clays. Rate constants for relevant homogeneous redox reactions are largely unknown, while subsurface low-temperature systems are often in redox disequilibrium. Reaction kinetics is also strongly coupled to the available reactive surface areas of potential reactant phases. Finally, nucleation mechanisms are often poorly understood, thus preventing adequate description of conditions leading to supersaturation and leading to inappropriate kinetic descriptions of the processes involved.

Third, while ion exchange and surface complexation models are commonly used in reactive transport models, data needed to support these models are generally not available. Furthermore, the available data are limited to primarily 25 C for conditions of local chemical equilibrium. Also, surface complexation models often suffer from the fact that they do not conserve charge when combined with transport processes.

Finally, chemical reactions sometimes alter the physical properties of the medium. Reactions of primary importance are mineral precipitation and dissolution and biomass formation and destruction. Such changes are difficult to quantify and usually require phenomenological formulations based on experiments. Typical coupled processes include changes in porosity and permeability, surface armoring, and changes in cation exchange capacity and surface site density.

Research Priorities: Chemical Processes	
2004	▪ Extend solution chemistry models to higher temperatures and ionic strengths (e.g., Pitzer equations) relevant to DOE contamination problems. ▪ Improve the understanding of kinetic rate mechanisms and effective surface areas, in order to develop improved rate laws. ▪ Determine if significant differences exist for measurement of cation exchange capacity and surface site density between in-situ field measurements of undisturbed samples and laboratory batch measurements involving disturbed media. Determine the validity of using batch Kd measurements to estimate retardation in variably saturated flowing systems. ▪ Initiate investigations into the effects of dissolution and precipitation on porosity, pore structure and permeability; initiate studies into nucleation controls that may bias dissolution and/or precipitation to particular pore environments.
2010	▪ Investigate mineral reactions in contact with a thin liquid film which may have different properties compared to the bulk fluid (e.g., for extreme evaporation conditions that may favor the formation of evaporite mineral phases). ▪ Investigate mineral reactions in unsaturated systems having variable gas phase chemistries (e.g., low and high CO_2 or O_2). ▪ Develop rate laws for glass dissolution and solid solutions that cover more extended conditions typical of natural and contaminant-affected flow fields. Improve models of nucleation processes. ▪ Determine the kinetics (and reversibility) of ion exchange and surface complexation reactions on mineral surfaces and colloids. Resolve the importance of lack of charge conservation in surface complexation models when combined with transport, possibly including streaming potentials in the formulation.

	▪ Develop a functional description for porosity/permeability evolution reflecting chemical effects.
2025	▪ Conduct experimental studies to develop appropriate databases of reaction rate constants applicable to refined reaction rate laws. ▪ Couple ion exchange and surface complexation processes to mineral precipitation and dissolution.

2.1.3 Biological Properties and Processes

Microorganisms are directly and indirectly responsible for numerous processes that influence the mobility and persistence of contaminants in the vadose zone. Many organic pollutants are utilized by microorganisms as electron donors or acceptors, and carbon sources. Many metals and radionuclides are metabolically transformed into chemical species that are more or less susceptible to long distance migration. In addition, microorganisms have a profound influence on their immediate environment and can alter porosity, surface characteristics, pH, and redox, which in turn affects contaminant transport.

In contrast to the saturated zone for which a relatively large microbiological knowledge base exists, much less information is available on the distribution, rate, and controls of vadose zone microbial processes. The vadose environment poses a number of challenges to quantifying microbial processes. Concentrations and fluxes of carbon and other nutrients are often very low and heterogeneous in their distributions. Inputs of nutrients to the vadose zone at many DOE sites via rainfall or other means are minimal and intermittent, and often localized to fractures and regions of preferential flow. Thus, microorganisms may have to survive long periods of dormancy, yet be prepared to take immediate advantage of transient and often brief influxes of nutrients. All of this reinforces the fact that microbial activity is very discontinuous and patchy in both time and space. Transport and colonization of microorganisms is another important issue. Microbes and contaminants are generally not co-located and a major challenge of engineered bioremediation is the delivery of microorganisms to sites of contamination.

Though many of the mechanisms for microbial interaction with a few specific contaminants are known, much remains to be learned about parameters that control the rate of transformations of radionuclides and mixtures of contaminants, as well as microbial processes in general under extreme conditions. In addition, it is not well understood how the unique hydrological features of the vadose zone impact metabolism, growth, and transport processes and their distribution. An understanding of these interactions and processes is required for reliable predictions of contaminant transport (enhanced transport and natural attenuation) and the potential for successful engineered bioremediation.

Major research goals to be accomplished are (1) understanding the mechanisms of microbial-contaminant interactions, including biodegradation and transformation, mobilization, complexation, and precipitation, for inorganics and contaminant mixtures in contact with vadose zone materials, (2) carrying out field and laboratory experiments and characterization to develop mathematical descriptions of key microbial processes and their linkage to spatially variable hydrogeologic and geochemical processes, and (3) application of emerging measurement and sampling technologies for understanding in-situ rates and scaling of microbial populations/activity in heterogeneous systems to scales relevant for field-scale modeling. These are discussed further in Section 3.2.3.

Research Priorities: Biological Processes	
2004	▪ Quantify rates of microbial processes affecting inorganics, chelate-inorganic complexes, inorganic complexes, organic contaminants, and identify factors controlling those rates. ▪ Understand and predict how contaminant bioavailability affects, and is affected by, microbial processes. ▪ Characterize microbial-mediated corrosion and biodeterioration of materials used to contain DOE contaminants. ▪ Develop and apply rapid statistical and classification methods for handling large data sets generated from the characterization of microbial communities.
2010	▪ Characterize and determine factors governing microbial diversity in the vadose zone. Determine relationships between diversity and community response to contaminants. ▪ Characterize microbial transport in different natural porous media relevant for DOE sites, particularly transport through unsaturated fine and coarse sands and fractured rock, as a function of water saturation and solution chemistry. ▪ Improve the conceptual and mathematical characterization of interactions between microorganisms and contaminants (e.g., biodegradation, immobilization, transformation), microbial and physical/chemical processes (coupled processes), and incorporate these equations into reactive transport models. ▪ Understand how mixtures of contaminants in DOE wastes affect microbial activity with respect to flow and transport of specific contaminants. Identify conditions when toxicity of radionuclides, metals, or organics prevents removal of readily degradable organic contaminants. Predict the rate of biodegradation of particular contaminants as a function of the type and concentration of other contaminants present. ▪ Understand the potential roles of siderophore, surfactant, and chelate production by microbial communities in the vadose zone, including the plant root-microbial community interface, on contaminant transformations. ▪ Elucidate pore-scale interactions between microorganisms & contaminants, including studies at the microscopic level, interfaces (solid/liquid, liquid/gas, liquid/liquid, solid/gas, and along textural discontinuities), & biofilms, under undisturbed conditions. ▪ Determine the importance of microbial attachment/detachment, biofilms and microbial associations with colloids with respect to contaminant transport and preferential flow.
2025	▪ Predict in situ rates of microbial biotransformation of contaminants at DOE sites as a function of site and environmental conditions. ▪ Incorporate information about the composition of vadose zone microbial communities into pollutant fate and transport models. ▪ Predict transport of native and introduced microorganisms at DOE waste sites. ▪ Describe and predict the spatial distribution of microbial biomass, activity, and community composition in the vadose zone with respect to contaminant distribution and predict how this distribution is affected by water inputs, contaminant fluxes, and duration of exposure to contaminants.

2.1.4 Colloidal Properties and Processes

Often, many contaminants such as radionuclides and metals are strongly sorbed by soil and sediments, thus limiting their transport in the subsurface. In some instances, however, strongly sorbing contaminants have been found at locations far beyond what would have been anticipated based on current knowledge and solute transport models. It is hypothesized that contaminants attach to small particles, called colloids, which may move relatively unhindered through the subsurface. Colloids with sizes between 1 nm and 1 ☞m can under certain conditions move large distances through a porous medium. Considerable advances have been made in understanding colloidal processes and transport in well-defined systems such as water-saturated sand filters. However, key processes including colloid formation, mobilization, and deposition in different types of natural porous media are still not well understood. While colloidal particles are naturally present in most soils, sediments and rocks, they do not necessarily form stable suspensions, and are therefore not very mobile. However, when certain system parameters are changed, such as the water flow rate or the solution chemistry, colloidal particles may become mobile and act as carriers for contaminants.

Microorganisms are a special class of colloidal particles. While their own fate and transport is strongly affected by their surface properties, microorganisms, like other colloids, can also actively affect organic and inorganic contaminants in the subsurface. For instance, microbial precipitation of soluble radionuclides and metals within cells or at cell surfaces can substantially alter the transport properties of these contaminants.

The specific mechanisms of colloid interactions with subsurface materials and the transport of colloids in natural soils, sediments, and rocks are not well understood. To quantitatively describe and predict the role and significance of colloids in contaminant transport, major research goals are (1) understanding mechanisms of colloid-contaminant and colloid-sediment interactions, including colloid formation, mobilization, and deposition, and (2) modeling colloid transport and colloid-facilitated transport in unsaturated soils, sediments, and fractured rocks.

Knowledge of colloidal processes in the subsurface improves the accuracy and reliability of predictions of short- and long-term contaminant fate and transport. Also, natural augmentation or artificial introduction of colloidal particles, such as microorganisms, into the subsurface is a potentially promising new remediation technology for many types of contaminants. Long-term stewardship of waste sites and the development of appropriate remediation strategies require detailed mechanistic understanding of colloidal processes in the subsurface.

Research Priorities: Colloidal Processes	
2004	▪ Consider mobile colloids in sampling and analyses protocols in field and monitoring studies. ▪ Develop new sampling techniques for *in situ* measurements of colloidal particles in pore water.
2010	▪ Quantify effect of preferential flow on colloid transport. Colloids may be mobilized at high flow rates (e.g. flow in fractures, instable flow, macropore flow), and as such preferential-flow phenomena may not only accelerate colloid transport but even lead

	to colloid mobilization. ▪ Understand relevance of colloid-facilitated transport under transient flow conditions, including wetting-drying and infiltration processes. ▪ Quantify potential for *in situ* colloid formation and mobilization under conditions relevant for DOE contamination sites, particularly in presence of extreme chemical conditions that can lead to dissolution and precipitation of soil minerals. ▪ Evaluate specific issues regarding microbial colloids. Research is needed on the effects of changing contaminant flux, nutrient injection during engineered bioremediation, and cell to cell communication (quorem sensing) in biofilms and other cell assemblages, on the production and behavior of microbial colloids. ▪ Characterize colloid-contaminant interactions as a function of solution chemistry and water saturation (this is needed at the microscopic level and the macroscopic level).
2025	▪ Model colloid transport and colloid-facilitated transport in unsaturated soils, sediments, and fractured rocks. ▪ Characterize colloid transport in different natural porous media relevant for DOE sites, especially colloidal transport through unsaturated fine and coarse sands and fractured rock, as a function of water saturation and solution chemistry. Colloid interactions with the solid-liquid as well as the liquid-gas interfaces need to be quantified. ▪ Improve conceptual and mathematical characterization of colloid-contaminant-soil interactions and colloid-facilitated transport and incorporation into reactive transport models.

2.1.5 Multiphase Flow and Transport Processes

Multiphase flow and transport of organic and inorganic contaminants in the vadose zone poses serious scientific and technical challenges. The vadose zone always contains multiple fluid phases through which contaminants may move, as well as associated interfaces for partitioning between phases. For example when air, water, nonaqueous phase liquids (NAPLs), and an immobile solid phase are present, three mobile phases may occur with as many as six interfaces for contaminant interphase mass transfer. The remediation of contaminated sites is ultimately controlled by these interphase mass transfer rates. Furthermore, contaminant loading rates in the saturated zone are controlled by migration and transport processes in the vadose zone.

Over the past several decades research has focused on quantifying flow and transport processes using idealized fluids and porous media. This research has greatly improved overall knowledge and the ability to predict multiphase flow and transport. However, additional research is necessary to quantify the effects of subsurface physical and biogeochemical heterogeneity on contaminant fate and persistence. Upscaling procedures to apply laboratory-derived constitutive properties and parameters to the field scale are especially important, including mass transfer and biological parameters that are typically measured at the soil column and microcosm scales. Unstable (e.g., fingering) and preferential flow of aqueous and NAPL phases is another important issue because of its potential to enhance contaminant mobility in the subsurface.

Additional difficulties arise when sites contain complex mixtures of inorganic and organic compounds such as heavy metals, radionuclides, organic solvents, and complexing agents. Interactions among aqueous phase contaminants are known to influence their geochemical behavior and transport. Very little research has explored and quantified the fate and persistence in the vadose zone of contaminant mixtures that may involve complex interactions among phases, interfaces, and chemical components. These interactions will likely influence multiphase flow and transport processes, as well as bioavailability of contaminants. Related questions must include, effects of chelating agents and NAPL phases on heavy metal and radionuclide transport, differences in the transport of complex NAPL mixtures versus pure NAPLs, the effect of aqueous chemistry on NAPL migration and transport, and the potential adverse and synergistic interactions among contaminants in mixtures during remediation operations. Understanding the nature of the contaminant association increases the probability of developing efficient remediation strategies that treat the contaminants simultaneously.

As discussed in more detail in Section 3.0, a combination of well-designed laboratory, numerical, and field scale studies are required to test specific hypotheses related to multiphase fluids. Multidisciplinary teams are needed to effectively address these complex issues. Collaboration among disciplines should improve descriptions of multiphase flow and transport processes in complex vadose zone environments, thus reducing uncertainty and ultimately providing specific tools for the design and implementation of cost-effective remediation and containment operations.

Research Priorities: Multiphase Flow and Transport Processes	
2004	▪ Explore the effects of subsurface heterogeneity (physical, chemical, and biological) on multiphase flow and transport processes. ▪ Develop upscaling procedures for laboratory-derived multiphase constitutive properties and parameters. ▪ Devise process-based models to describe unstable and preferential flow of aqueous and NAPL phases.
2010	▪ Measure flow and transport properties of key contaminant mixtures. ▪ Refine theory and numerical models describing flow and transport of contaminant mixtures. ▪ Conduct multiphase flow and NAPL transport experiments for representative soils and contaminants to elucidate real-world problems and data limitations. ▪ Design, implement and analyze controlled tests of complex contaminant mixtures in highly heterogeneous systems.
2025	▪ Refine remediation strategies for site specific contaminant mixtures. ▪ Use numerical models to assess uncertainty in flow and transport of complex contaminant mixtures at DOE sites, and to support decisions with regard to remediation and containment strategies.

2.1.6 Chaotic and Unstable Processes

Field observations of liquid flow and chemical transport in both porous and fractured unsaturated media often show unstable, random spatial and temporal fluctuations in the moisture content, pressure, flow rate, temperature, and concentration of chemicals. These fluctuations can result in rapid preferential flow and transport through the vadose zone to the groundwater table. Despite numerous studies of preferential flow, the causes of flow instability are still not completely known. It is apparent that flow in structured, macroporous soils and fractured rocks is significantly influenced by abrupt changes in media physical properties (e.g., rough to smooth fractures, small to large pores) and an interplay between gravitational and capillary forces. These interactions generate liquid fragmentation (bridges, fingers, rivulets), intermittent flow, and extreme sensitivity to small changes in boundary conditions and variations in media geometry, which may not exist in homogenous porous media. Other examples of processes leading to chaos are discontinuous processes in the liquid behavior in porous media such as Haines jumps, flow-induced nonlinear effects in thin liquid films, drainage by cavitation, nucleation and mineral precipitation within pore spaces, density driven flow, dripping water in fractures and caves, fracture-matrix interaction processes, and thermal effects. The combination of several nonlinear physical processes involving both deterministic-chaotic and random flow and transport processes may cause spatial and temporal fluctuations of flow parameters. Continuum representation models have limited utility for inherently chaotic systems because of the need for different governing equations describing intermittent flows in different parts of the system and at different times. To better understand the physics of processes leading to chaos, special experimental and modeling investigations are needed. Models are needed to describe coupled highly nonlinear processes of flow and mass transport in variably saturated media. It is also important to identify criteria (such as Bond and capillary numbers) that can be used to determine the onset of chaos.

Chaotic dynamics is one of several new fields in modern science attempting to better understand order and pattern in nature. In particular, it may improve our understanding of the physics of chaotic flow processes in variably saturated porous and fractured media, which are described with both deterministic and stochastic models. The knowledge of the presence of deterministic chaotic processes is important because the long-term predictability of a deterministic-chaotic dynamic system is limited. For such a system, one can provide precise short-term predictions using deterministic-chaotic models and determine only a range of possible long-term predictions using stochastic models.

Research Priorities: Chaotic and Unstable Processes	
2004	▪ Review literature related to physical, chemical, and biological processes for which the presence of chaos was established providing comparative analyses of these processes and those occurring in the vadose zone. ▪ Analyze factors leading to flow and chemical transport instability and chaos based on a series of bench-scale and field investigations. ▪ Develop a series of models describing phenomena of deterministic chaos in

	unsaturated fractured rocks. ▪ Develop criteria to identify the onset of chaos. ▪ Identify applications of chaos theory in vadose zone hydrology
2010	▪ Design and perform field-scale infiltration tests with tracers at several DOE sites with heterogeneous soils and fractured rocks to identify conditions for which chaotic processes are important. ▪ Develop a new generation of mathematical and numerical models based on theory of chaotic dynamics to describe unstable, chaotic flow processes in soils and fractured rocks. ▪ Develop software for the determination of main diagnostic parameters of chaos using the reconstruction of the system phase-space from scalar data.
2025	▪ Implement models of unstable, chaotic flow into other deterministic and stochastic models used for predicting flow and transport and designing remediation activities.

2.2 Possessing Adequate Data

All scientific endeavors require data –to validate hypotheses, to create models, to confirm or alter theories, and to develop and verify simulations of future events. Characterization and monitoring methods provide data that are crucial to understanding vadose zone processes, and in turn the data are essential for making good decisions with regard to DOE site remediation and long-term stewardship. Characterization of vadose zone processes and heterogeneities provides the data required to develop accurate simulations with the appropriate spatial resolution. Monitoring contaminant migration confirms the accuracy of simulations, as well as acts as a trigger for remedial action.

The long-term vision for characterization and monitoring is to generate adequate data for a four-dimensional description (three dimensions plus time) of the relevant processes and parameters in the vadose zone. This vision depends upon integration of measurement and interpretation across several disciplines, such as biology, geology, geochemistry, hydrogeology, geophysics, and geomechanics. In addition to integrating the different disciplines, this vision will require a massively parallel computer initiative similar to the Accelerated Strategic Computing Initiative (ASCI). A state-of-the-art massively parallel machine (an ASCI Green Machine?) will have to be available for priority use by university and national lab researchers working on scientific and technological solutions for characterizing and monitoring of DOE contaminated sites.

To achieve a four-dimensional description of the vadose zone, advances are required in the tools, techniques, and approaches used by DOE and the scientific community. Research and advancement needs are discussed below in seven broad areas: (1) the Geological, Chemical, Biological, Hydrological (GCBH) Framework, (2) Chemical Properties and Processes, (3) Biological Properties and Processes, (4) Hydrogeological Properties and Processes, (5) Network Design and System Optimization, (6) Sensors & Instrumentation, and (7) Integration Issues. In the first of these categories (GCBH Framework) the issues involved in understanding basic processes are discussed from the perspective of characterization and monitoring. Areas two through five address state variables and properties that need to be measured. Areas six and seven discuss important aspects of how these measurements should be made.

2.2.1 Geological, Chemical, Biological, Hydrological (GCBH) Framework

A significant number of subsurface environmental problems involve fluid flow and solute transport in the vadose zone. A key component in assessing contamination hazards and designing remedial actions is the development of flow and transport models. These models are developed to work within a specific physical framework that accounts for appropriate processes at the appropriate scale. The physical framework must include the relevant geological, chemical, biological, and hydrological parameters and processes in 3-dimensional space, while taking into account changes in these processes and parameters over time.

Framework development also requires researchers to use a common approach for assessing spatial heterogeneity in media properties at multiple scales (molecular to regional). Although this is commonly accepted in any characterization program, initial evaluation of model uncertainty and data needs will identify any limitations or data gaps for a particular site. By identifying data gaps, critical process understanding (or the recognition of a lack of understanding) can be arrived at early in the characterization program depending on what the relevant processes are for a given site (e.g. fracture dominated flow or matrix dominated flow). The development of the framework must be conducted as an interactive process in concert with numerical modeling and experimental analysis. A more in-depth discussion of the necessary interaction and integration amongst understanding of basic processes, model development and validation, and characterization and modeling capabilities is provided in Section 3.0.

Long term goals are to: 1) develop a classification scheme for the hydrogeologic settings of major DOE sites, 2) use or develop geologic/geomorphic understanding to interpolate point data to 3-dimensional space, 3) develop a working relationship amongst depositional, structural and igneous features to the relevant parameters and process and, 4) develop hypotheses with respect to critical features and implement methods to determine critical features for important flow regimes.

Research Priorities: Understanding the GCBH Framework	
2004	▪ Tabulate known features for individual sites and start using the geologic/geomorphic understanding to develop 3-dimensional framework models.
2010	▪ Develop prototypical numerical models that incorporate the 3-dimensional framework model with the relevant parameters and processes in place.
2025	▪ Use the models and all existing data and observations to determine the critical features, parameters and processes for the sites. In addition, simulations of flow and transport should be consistent with all existing data and observations.

2.2.2 Chemical Properties and Processes

The role of chemical characterization and monitoring in vadose zone understanding includes the spatial and temporal description of chemical fate and transport. From this description in space and time, the concentration and extent of contaminants and the identification of appropriate conceptual models can be determined.

Concentration and Extent of Contaminants

The spatial distribution and concentration of contaminants is the primary issue in most environmental contamination problems faced by DOE. Knowledge of the extent and concentration of contaminants is essential to: Establishing the existence and magnitude of a contamination problem at a particular site; Determining whether contaminants have moved offsite and/or to environmental receptor points; Determining likely sources and rates of transport; Providing a quantitative basis for assessing health and environmental risks; Guiding remediation decisions; Evaluating/demonstrating the effectiveness of remedial efforts; Providing a well-defined initial condition for simulation of future contaminant migration and fate; and Increasing our understanding of subsurface transport processes.

The concentration and extent of contamination is presently determined using invasive sample collection methods (typically boreholes) and laboratory analysis of samples. In situ probes monitor some contaminants with near-real-time analysis and transmit data using datalogger technology. Some contaminants (e.g., radioactive constituents) can be observed using borehole geophysical methods. In most cases, concentrations are measured on a discrete time interval (e.g., quarterly) and concentrations are measured at only a limited number of points in 3-D space.

To improve our current methods the first task is to select technologies that can identify the characteristics of individual contaminant species. Candidate technologies include spectral IP, NMR, spectral EM (including radar), neutron logging technology, and chemical sniffing methods. Second, the relationship of the measurements to the contaminant species must be established. Through lab analysis, chemical species can be isolated by their geophysical or geochemical responses. Third, the sensitivity limits of the measurement to the contaminant species and its concentration must be determined. Fourth, prototype tools or combinations of tools suitable for a restricted environment need to be deployed. This progressive approach will probably be applied first to logging technology. Lastly, these technologies all must be extended to full site investigations.

Source Identification and Monitoring

Historically, mixed wastes have been placed in underground storage units or disposal facilities. Many of the remediation alternatives for existing contaminated sites or decommissioned facilities will leave residual wastes in place. Waste disposal alternatives include encapsulation of waste containers in sediments, concrete, and other engineered barriers, and disposal in underground tanks and cribs. As wastes are disposed of at or near the surface, these residual wastes have the potential to release contaminants that migrate through the vadose zone. Wastes of interest include organics, heavy metals, and radionuclides disposed of as either solids or liquids. These wastes can be transported in the gas phase, aqueous phase, or as non-aqueous liquids. Non-aqueous liquids residing in the vadose zone can also serve as long-term sources for groundwater contamination. Methods for non-invasive monitoring of these potential sources are important, because once released to the heterogeneous subsurface, detection of contaminant migration is extremely problematic and very expensive. Non-invasive identification of subsurface contamination sources, such as barrels and other buried wastes, could also be invaluable in planning and executing safe and cost-effective remediation.

To date, there have been limited studies of geophysical methods of characterizing buried waste. These methods are similar to those used to identify other subsurface cultural features and are currently limited in their ability to distinguish various types of waste. Usually, identification of contamination sources relies on invasive sampling, incomplete disposal records, and modeling. There are currently no techniques for monitoring real-time changes in source contaminant configuration (e.g., migration of contaminants in the vadose zone below disposal cribs or tanks).

Fate and Transport Monitoring and Conceptual Model Identification

Long-term monitoring of vadose zone contamination must assure contaminant isolation as well as detect unintended releases. This should be done in an iterative fashion by continually updating site conceptual models with new monitoring data. Monitoring should not begin until a conceptual model is in place, and monitoring data should refine or test the conceptual model. It is essential to explicitly link monitoring to contaminant fate models before monitoring starts, so that subsequent monitoring efforts achieve their purpose. To facilitate this linkage, a portfolio of "fate" models that are grouped by contaminant type should be developed.

Research Priorities: Chemical Properties and Processes	
2004	▪ Build conceptual models for attenuation of DOE contaminants on field data of specific contaminant plumes. At least 2 to 3 conceptual models are expected to describe the attenuation of particular contaminants. These will initially be catalogued and periodically updated in the light of ongoing research (2000 - 2005) ▪ Determine if various contaminants are present in soils from surface or borehole measurements. May need to couple field results with lab analysis to verify the association. ▪ Delineate in situ burial/disposal configurations (barrels, drums, boxes, and subsurface structures) distinguishing target contaminants from cultural features and soils.
2010	▪ Examine and compile field measurements from within and outside the DOE complex to provide broad-based technical support for the portfolio of "fate" modes. Characteristic transport distances, chemical controls on attenuation, and hydrologic factors will be determined on a site-specific basis but will be used collectively to update the conceptual model portfolio. Two parallel efforts will be carried on. 1. Cataloguing of contaminant-specific monitoring needs; and 2. Specific identification of "standard" monitoring approaches that cannot conceivably update or refine the cast of conceptual models for the specific contaminant. The latter effort is important because a large fraction of existing monitoring data is orphan data (will never be used). Minimize data collection that is likely to be orphaned (2005 - 2015).

	▪ Determine the concentration of the most abundant species within an order of magnitude. This probably does not apply to very small concentrations (PPM). ▪ Develop a cost-effective real-time monitoring and data analysis system to provide early warning of contaminant discharges to the vadose zone.
2025	▪ Coordinate a DOE-wide effort to collect long-term monitoring lessons learned. At this time there should be enough long-term monitoring data gathered to condense the conceptual model catalogue and to standardize monitoring approaches and to institutionalize a reflexive and iterative linking between monitoring and model refinement (2015 - 2025). ▪ Isolate contaminated volumes and determine the concentration of abundant chemical species within 10 or 20 percent. ▪ Quantify the volume and spatial distribution of resident contaminants at or near source discharge points, for metals, radionuclides, and organics.

2.2.3 Biological Properties and Processes

Nearly all state-of-the-art microbiological characterization methods currently require invasive sample collection and most require subsequent laboratory analysis (versus downhole data collection). These requirements result in several technical problems: high cost; inability to measure rates in situ; spatial heterogeneity and temporal dynamics (small and local variations in physical and chemical properties can result in large differences in the microbiology); general lack of joint measurements; and inadequate scale of measurements.

Biological processes are important to invasive vadose zone characterization and monitoring in two ways. First, microorganisms are involved in many processes affecting contaminant transport and transformation, and their effects must be quantitatively understood. Second, biological processes, biomolecules, and biomaterials can be used as components in instruments and sensors for characterization and monitoring the vadose zone.

Non-invasive techniques have been used only in an exploratory fashion to observe the distribution and activity of microbes in the subsurface. Preliminary experiments in the lab and in limited field tests have shown that increased concentrations of microbes and/or movement may produce electrical signals, which may be picked up from the surface or in near-by boreholes. Current thought is that the self-potential effect may be responsible for the signals. In general, however, invasive sampling and laboratory analysis is required to determine subsurface microbial activity. This greatly limits our ability to resolve spatially heterogeneous and temporally varying microbial distributions.

Three major types of microbial data are needed for site assessment and site monitoring (bioavailability, in situ rates, and community composition and activity). Forged by the intersection of biology, microfluidics, microelectronics, and micro-optics, biotechnological advancements pertinent to all three types of data are occurring rapidly in the private sector. However, we must invest in research to adapt and apply these advancements to vadose zone science and stewardship.

Bioavailability. It is important to know what concentration or mass of a contaminant is immediately available for microbial transformation or is capable of toxic effects on organisms. We also need ways to measure the fraction that may eventually become available, as well as improved in situ and ex situ approaches.

In situ rate. We need approaches for measuring in situ rates at the intermediate (0.1 to 10 m^3) and miniature (mm^3 to cm^3) interrogation volume scales. For field applications, approaches similar to the in situ respiration test (to measure use of injected oxygen to estimate petroleum hydrocarbon degradation) using gases and/or solutes are needed. Challenges to successfully utilizing such approaches include the development of instrument systems that minimize changes in moisture status and reduce the creation of near-well-bore artifacts during the assay, and sufficient analytical sensitivity for often very slow in situ processes.

Community composition and activity. Rapid advances are occurring in high-resolution signature lipid biomarkers, DNA chips, and other nucleic acid methods for characterizing microbial community structure, diversity, and activity. These measurements are important for improving our understanding of vadose zone microbiological processes and linking communities to functions. However, the per sample cost is high.

Research Priorities: Biological Properties and Processes	
2004	▪ Develop improved bioavailability sensors/assays. ▪ Develop sensors/systems for in situ rate measurements. ▪ Develop chemically reactive contact foils and films for in situ rates and/or potential activity. ▪ Develop sample-to-answer analytical systems for community composition and activity. ▪ Develop spectroscopy- and synchrotron-based inferences of microbial activity. ▪ Laboratory and modeling of the bio-electric level as a function of different soil types and microbes (aerobic vs. anaerobic, variety of metal and organic reducing microbes).
2010	▪ Application of in situ bioavailability sensors. ▪ Application of sensors/systems for in situ rate measurements. ▪ Application of inexpensive sample-to-answer analytical systems for community composition and activity. ▪ Instrumentation development to detect lower levels of activity. ▪ Intermediate and field scale validation tests.
2025	▪ Develop economical analysis of spatial distribution and temporal dynamics. ▪ Extend nondestructive joint measurement of biologic, geochemical, physical, and hydrologic properties or processes to intact cores, and possibly the subsurface. ▪ Develop proxy measurements for microbiological properties and processes.

2.2.4 Hydrogeological Properties and Processes

The role of hydrologic characterization and monitoring in vadose zone understanding includes the measurement of state variables such as moisture content, matric potential, temperature, and the description of flow and migration (perturbed and unperturbed). Flow and migration include the determination of hydrologic parameters such as permeability and porosity in time and space, the flow architecture including fractures, and the suite of methods (e.g., tracers) used to map the subsurface.

Moisture Content, Matric Potential, and Temperature

Methods for assessing state variables are primarily invasive and have not evolved much in several decades. This is a key area for development, including: improving robustness; easing emplacement costs and containment system concerns; and simplifying data communication, fidelity and man-power-requirements.

Among the most important state variables requiring characterization in the vadose zone are:

- Water content¾ the mass of water held in a soil is often reported as percent saturation. Water contents are needed in order to make predictions of contaminant transport through the vadose zone and for mass balance calculations.

- Soil water potential or matric potential¾ the energy status of water in soil or other material resulting from its presence in the gravitational force field, capillary and surface forces, and from the presence of mineral salts. Matric potentials are needed to determine gradients for water flow.

- Temperature of the soil¾ important for many processes such as microbial activity, water and vapor movement and to some degree contaminant transport. Many biological and chemical processes depend on temperature.

Distribution of Flow and Migration (Perturbed and Unperturbed)

Mapping water distribution and tracking water migration in the vadose zone are among the most important tasks for noninvasive monitoring. Water is the chief vehicle for transporting other chemical species, and it is an important solvent in itself. Tracking water movement may also reveal fundamental formation parameters such as porosity and permeability.

Presently, water saturation within the vadose zone is poorly determined from geophysical measurement; greater success has been achieved in time lapse studies of water movement. Both electrical and seismic techniques typically have good sensitivity to water saturation in the vadose zone, but there are many other competing factors that make isolating the effect of water saturation difficult. This suggests that an approach using several independent measurements would be effective. Other promising technologies include spectral IP, borehole and ground penetrating radar, spectral inductive EM, and microgravity. All of these have shown a good sensitivity to water saturation and flow in some conditions. Ideally we would like to map water distribution and track flow within a volume of interest in an arbitrary geology using cost-effective noninvasive technology.

Tracer (Natural and Induced) Migration and Interaction

Characterization and modeling of subsurface flow and transport is a critical issue to evaluate current distribution of subsurface contaminants and to predict future migration. Natural and anthropogenic tracers can be used to determine directions and rates of transport, assess flow processes, and validate numerical models. The subsurface distribution of contaminants can also be considered a large-scale applied tracer experiment; however, it is often difficult to reconstruct location and timing of inputs. The challenge is to optimize the use of tracers and extract as much information about the subsurface as possible while providing data to complement information from other approaches to provide a consistent model of the system.

Research Priorities: Hydrogeological Properties and Processes	
2004	▪ Improve methods for moisture content, matric potential, and temperature ▪ Track a broad waterfront in a 10m X 10m area, at depths up to 100m, to a precision of 10%. ▪ Inverse process of using tracers to estimate fluxes and ages of pore water; propagation of uncertainties to obtain uncertainties in calculated fluxes and ages (2004 - 2025). ▪ Use of a variety of tracers for locating contaminants; importance of surface boundary conditions; and point estimates of fluxes and ages (2004 - 2025).
2010	▪ Resolve the same front to 5% over a 50m X 50m area.
2025	▪ Identify a 3D pulse of water (2m X 2m) to an accuracy of about 2% at depths of up to 100m. To achieve this goal we need both an improved imaging technology and better integration with hydrological and geological information.

Quantification of Flow in Fractures

Geophysics Focused on Fractures. Fractures often serve as preferred pathways for migration of contaminants. In the vadose zone, fractures almost always represent zones of permeability that are different from normal, unbroken rock or undisturbed, unconsolidated materials. Consequently, knowledge of the location and physical properties of fractures is necessary for evaluation of flow regimes, modeling calculations, and remediation planning. The spatial distribution of fractures, fracture trace length, and the connectivity distribution of fractures are often inferred using cores, outcrop data, borehole cameras, borehole temperature and flow meters, borehole acoustic televiewers, hydraulic tests, and seismic methods.

We need to capture the spatial variations of the fractures or fracture systems in an efficient manner that is not dependent on local borehole stress conditions. A crucial component for fracture characterization is distinguishing between the presence of any/all fracture(s) and the subset of fractures that serve as conduits or barriers for fluid migration. Crosshole geophysical data have already been used to identify the presence of fractures as well as water-conducting fracture pathways. Crosshole geophysical data may also provide valuable information about the presence and nature of fractures away from the borehole wall. Surface geophysical techniques include the use of P-wave reflection techniques to detect shallow faults with offsets as small as two or three meters. A potentially useful seismic method not used in petroleum exploration seismology is diffraction enhancement; detection of faults could be improved by enhancing diffractions rather than removing them, which is characteristic of migration processing. Research on relatively high frequency (10-100 Hz) surface waves in multi-channel analysis of surface waves suggest that faults and joints might be routinely detectable with surface waves.

Some general issues associated with the geophysical fracture characterization approach include fracture imaging and sensor development, understanding the geophysical responses to fractures, and integrated interpretation to address scaling issues.

Flow in Fractures. The ability to detect and determine the nature of fluid in fractures from surface or borehole measurements is important in many environments, but especially in hard rock geology. In hard rock environments, fluid contaminants are stored and migrate principally in the fracture network. Currently, no surface methods can reliably detect fluid in fractures in the vadose zone. There have been reports successes of crosshole radar measurements detecting injected tracer fluid in fractures, but there are no cases where in situ fluid in fractures has been detected or imaged. The best candidate technologies appear to be seismic techniques and radar, which provide both the range and resolution required. Other interesting possibilities include geochemical methods, electrical and electromagnetic techniques, and logging techniques.

The goal is to detect the presence of fluid in fractures and to quantify the amount in real time from the surface.

Research Priorities: Quantification of Flow in Fractures	
2004	▪ Detect the presence of fluid in fractures in some cases. ▪ Quantify the relationships among the most likely geophysical methods or combinations of methods that will provide fracture diagnostics
2010	▪ Detect contaminant fluid in fractures in most cases. ▪ Discover and quantify new relationships between surface geophysical measurements and fractures, so that fractures a few centimeters across at depths of 10 meters will be routinely mapped by geophysical measurements.
2025	▪ Detect and quantify contaminant fluid in fractures in real time from the surface or in boreholes. ▪ Have low-cost, automated and reliable geophysical techniques for mapping fractures immediately, at the field site, without delays for processing or analysis

2.2.5 Integration Issues

Many of the problems and initial cost of subsurface remediation comes from field site characterization. Three-dimensional information about the heterogeneous subsurface is needed in order to identify the key controls on flow and contaminant transport processes. It is widely recognized that natural heterogeneity and the large spatial variability of permeability predominantly control the flow field and hence the transport. Moreover, natural heterogeneity exhibits variability over a wide range of scales and is difficult to characterize using only one-dimensional borehole data. Geophysical data can complement direct borehole characterization data by providing multidimensional subsurface measurements in a minimally invasive manner. Crosshole geophysical methods are particularly promising characterization tools as they have the potential to provide high-resolution information about geophysical attributes in the intra-wellbore area. Our goal in environmental characterization is to use these high-resolution, multi-dimensional geophysical attribute images to infer key hydrogeological properties of the subsurface such as permeability (or saturated hydraulic conductivity), porosity, water content, and lithology or texture.

There are currently several obstacles that must be fully investigated prior to routine use of crosshole tomographic data for estimating hydrogeological properties. Some of these issues are briefly described below and the proposed research areas are summarized in the table following these descriptions.

Petrophysical Relationships

The ability to characterize petrophysical relationships (e.g. the relationship between dielectric constant as measured with a TDR probe to moisture content) permits the translation of geophysical attributes into estimates of hydrogeological parameters such as porosity and water content. Although much work has been dedicated to investigating such relationships for consolidated rocks under high pressures (conditions common for petroleum reservoirs) development of such relationships for unconsolidated, low pressure materials common to many near surface sites is still an infant topic of research. Currently, most investigations (at best) obtain a site-specific relationship between co-located geophysical attributes and hydrological parameters. There is lack of data and consensus on many issues including what kind of trend to expect (for example, does dielectric constant from radar tomography in general increase or decrease with increasing porosity/permeability?) and how grain sorting affects the geophysical response. Few attempts have been made to move beyond site-specific development of these relationships to fully understand the interaction between field-scale geophysical responses and the physical properties of the medium. Similarly, there has been little research on how to include multiple geophysical attributes in the relationship to reduce uncertainties, and how to handle non-uniqueness conditions sometimes inherent in petrophysical relationships.

Estimation Methodologies

Routine procedures must be developed to incorporate geophysical tomographic measurements with existing direct measurements from wellbores and with other data (geological information, surface geophysical data, chemical data, and biological data) in a rigorous manner. These methods also must provide better quantification of estimate uncertainty arising from measurement error, inversion error, and use of an estimated petrophysical relationship. We must understand how use of multiple, co-located data sets to reduce the ambiguity and uncertainty associated with parameter estimates. Some stochastic methodologies have been developed to handle some of these issues, but the methodologies would benefit from refinement and further testing and extension with scale issues in mind.

Inversion Approaches

Inversion approaches, such as those involving crosshole tomography, must be further developed and tested. This includes reaching consensus on appropriate data correction procedures prior to inversion, as well as more advanced approaches such as constrained inversion, stochastic inversion, and joint geophysical-geophysical or geophysical-hydrological inversions. We should investigate how to weigh the information content of the different geophysical data prior to joint inversion, how to invert data having different resolutions, and how to correctly provide uncertainty estimates corresponding to the inverted tomographic attributes.

Field Tests

The modeling process is comprised of the following four general steps: 1) develop conceptual model, 2) formulate mathematical model, 3) implement solution technique, and 4) perform model verification and testing. Errors in the conceptual model can lead to errors in predicting and understanding vadose zone flow and transport. Alternative conceptual models are proposed when lack of data or process understanding does not allow unique identification of a single model, site characterization defines a new feature or process, or model testing rejects a candidate conceptual model. A major barrier to identifying alternative conceptual models is the design and execution of experiments that discriminate among various models.

Scaling

Issues of scale must be investigated when developing petrophysical relationships, applying/developing estimation methodologies, and integrating different types of data. The large spatial variability of hydraulic properties in natural geologic systems over a wide range of scales renders estimation of hydraulic parameters and their spatial correlations difficult. Further compounding the problem is that the obtained measurement value is a function of the instrument support scale (volume of investigation), as well as the network scale (acquisition parameters/instrument spacing). Both the instrument and network scales vary amongst and even within geophysical methods. Thus, it is crucial to understand how these measurement-related scales impact the obtained value, and how to incorporate different types of measurements with each other. There should be general understanding as to how to scale measurements collected in the laboratory for use in the field, how to utilize petrophysical relationships developed in the lab with field data, and how to integrate different data that sample different volumes.

Research Priorities: Integration Issues	
2004	▪ Routinely develop petrophysical relationships for different environments, and start to understand how multiple data reduces estimation uncertainty and aids with issues of non-uniqueness between geophysical and hydrological parameters. A complex-wide database of representative geophysical, chemical, hydrological and geological parameters should be established to document distributions for different parameters as a function of their geological conditions. ▪ Become comfortable using stochastic estimation techniques that provide an estimate

	of the property of interest as well as information about its uncertainty. Develop better estimation techniques to facilitate routine incorporation of different types of data (GCBH) that have different sampling scales and different spatial variability. ▪ Develop automatic data picking and quality control approaches for crosshole tomographic methods. Also investigate the utility of constrained and joint inversion for improved estimation, and develop stochastic inversion procedures that yield distributions of possible geophysical attributes at each location. ▪ Design controlled field experiments distinguishing amongst conceptual models for better assessment of vadose zone flow and transport issues by providing a methodology for model testing, a more formal approach to model formulation, and a larger experience base of observing and measuring vadose zone flow and transport under controlled conditions (2004 - 2025). ▪ Conduct vadose zone field experiments to test and refine models and measurement techniques. This will include advances in instrumentation, tracers, biological properties, and emplacement technologies as well as mathematical and statistical bases for design. Scale issues, as relative importance of processes to be included in a conceptual model and parameter identification, are critical for field experiment design (2004 - 2025). ▪ Begin to test models by comparing model predictions to short (and eventually long term) observations (2004 - 2025). ▪ Recognize the importance of scale on characterization measurement and incorporate measurement support scale in estimates of hydrological properties and correlation lengths. ▪ Report scale parameters, as well as measured values and distributions to a complex-wide database on GCBH parameters as a function of geological environment. Vadose zone scientists should also by this time be comfortable interpreting their measured data using a hierarchical structure of organization that can exist in natural geological environments.
2010	▪ Understand at the mechanism of geophysical energy propagation within un- or loosely-consolidated, low pressure, granular porous material at the field tomographic scale, and what are the key influencing factors on the resulting geophysical signature. Understand the sensitivity of different geophysical tomographic methods to varied geological conditions, and know if multiple geophysical methods are cost-effective for certain investigations. ▪ Provide estimates of hydrogeological properties (e.g. water content, permeability and porosity) and their associated uncertainties and spatial correlation functions using different types of data such as crosshole tomographic. Estimates should be conditioned to direct measurements, and estimation will be made with attention to scale, using hierarchical spatial scale estimation procedures with multi-scaled data. ▪ Understand which geophysical method or combination of methods is most cost-effective for characterizing different geological environments or dynamic processes. ▪ Learn how to perform joint and constrained inversions that honor all data, understand how the additional data improves the estimate and decreases the error, and how to incorporate data with different measurement scales in the inversion process correctly. ▪ Investigate potential of collecting/inverting multi-scaled data to provide information about variabilities as a function of scale. ▪ Develop a quick and reliable way to assess the scale of the contamination problem relative to the scale of the hydrological heterogeneity understanding which technique

	or acquisition parameters are most appropriate for characterization of the key parameters that control flow and transport. ▪ Establish methods to incorporate different types of data to assess parameters over different spatial scales. ▪ Attempt to modify the acquisition parameters within a certain type of method to sample different scales of variability and understand how the measurements scale from one level of support to another.
2025	▪ Establish guidelines for different environments so quick field tests can determine which category the system under investigation falls into, and utilize the governing petrophysical relationship for that regime with the geophysical data in the estimation routine for at least first-pass estimations. ▪ Use tomographic methods to supply real-time, multi-dimensional (spatially and temporally), automatic interpretations of hydrogeological properties or changes in properties due to bacterial modification, environmental remediation, infiltration, and other dynamic processes. ▪ Apply the inversion process automatically to provide multi-dimensional, possibly multi-scale, and real-time estimates of hydrogeological properties at high resolution. ▪ Understand how to scale between laboratory- and field-based measurements, and incorporate a variety of differently scaled data to provide an integrated and hierarchical interpretation of hydrological properties

2.2.6 Network Design & System Optimization

Network Design

Subsurface characterization and monitoring for contaminants is difficult and expensive. The question is, how can we get the most information about subsurface contamination with the fewest boreholes, monitoring devices, etc? Or, how can we be sure that for the case of compliance monitoring the vadose zone monitoring system will indeed detect if there is a leak of contaminants from the engineered waste disposal site?

Subsurface characterization for waste plumes in the vadose zone is quite similar to detecting mineral deposits in geologic media. The statistical technique of kriging was developed for the mining industry to optimize the spacing for exploration boreholes. In the oil industry, pattern gridding and other techniques have been developed for locating petroleum reserves.

Monitoring network design has been used to optimize the placement of monitoring points for detection monitoring at disposal sites. However, many of the models used for designing a compliance monitoring system have been developed for groundwater monitoring. Little, if any work is ongoing for compliance monitoring in the vadose zone.

The state-of-practice needs to be reviewed in the area of subsurface monitoring system design; optimization methodology for subsurface characterization needs to be developed further; and compliance monitoring for the vadose zone should be encouraged.

Value of Data

Vadose zone characterization and monitoring technology is evolving rapidly. This evolution presents many options in developing and managing environmental sites for remediation or long-term stewardship. Among these options are 'high tech' parameter-measurement approaches, e.g., 3D and time-lapse 3D geophysical surveys, novel emplacement methods, and advanced sensors. Today's environmental management team must be able to use new technologies and methods that have been designed to provide better and more accurate information about the site in question. But how does the team evaluate the various options to maximize the environmental surety and minimize the economic impact of achieving environmental surety? How do they know whether a different approach might not yield a greater return or reduced risk? What data is required to optimally address a given situation, yet obey a restricted data acquisition budget? How does the team convince itself that novel and, perhaps, more expensive data collection, observation, and sampling well design options are worth the additional cost because the net return will be much greater?

At present, there are no objective means to evaluate reservoir technologies. Over time, the repeated application of a technology demonstrates a level of success that encourages further use, and eventually the new technology becomes standard practice. This 'Darwinian' approach to evaluating environmental technologies does succeed in the end to identify tools that benefit environmental management, but it also effectively denies access to the benefits of those tools for the vast majority of sites or projects that cannot afford the risk of 'experimenting' with new technologies. The overall goal should be a system (e.g., a Decision Support System) that a non-decision theory expert can use readily and rapidly to prioritize locations of characterization and monitoring activity and allocate discipline-specific manpower and resources.

Research Priorities: Network Design and System Optimization	
2004	▪ Numerical laboratory combined with highly instrumented field sites ▪ Define numerical experiments to assess the value of data ▪ Develop methods to determine sensitivity of parameters ▪ Reduction of uncertainty (affected by cost, data density, level)
2010	▪ Upgrade numerical laboratory and combine with field data ▪ Apply methods to field scale tests and experiments ▪ Develop methods to identify controlling parameters
2025	▪ Determine sensitivity for complex-wide sites ▪ Convey to endusers at what cost, data density and level can you reduce uncertainty

2.2.7 Sensors and Instrumentation

Improved Sensors, Drilling, Sampling and Sensor Emplacement

The nature of invasive vadose zone characterization and monitoring requires access to the subsurface through boreholes, trenches and other excavations. While this direct access enables more direct characterization at many scales, the direct access also raises issues that have created barriers for the use of invasive methods. These barriers include:

- Problems of Access – Some contaminated sites are "hot," therefore the exposure of personnel and equipment must be limited.
- Effects on Flow and Contaminant Transport – Drilling, direct-push and other methods create new pathways for flow and contaminant migration.
- Effects of Sampling - The direct access methods directly perturb the soil and rock, therefore, altering the processes and parameters of interest.
- Maintenance of Characterization and Monitoring Systems – Reliability is an issue for monitoring over long time periods.
- Data Communications - Longer-term characterization and monitoring requires new approaches to communicate data from the subsurface and to minimize or automate the data recording to selected intervals.
- Improved Sensors - Over the next 25 years, there will be a revolution in sensor devices. While the market for environmental restoration may not drive this new generation of sensor development, the environmental communities must find ways to influence the development of these sensors for new applications in order to leverage the anticipated growth of this technology.

Research and development of new sensors, improved emplacement of new and current sensors, and novel emplacement approaches are key to enabling invasive characterization and monitoring.

Instrumentation and Interpretation Needs – Geophysics

The major parameters of interest in characterization and monitoring are: lithology, soil type, porosity, mineralogy, fluid content and type, stress, displacement, pore pressure, groundwater velocity, and permeability (fluid and gas). Parameters that can be measured by geophysical methods that have an indirect or interpretable relationship to the required parameters for site characterization are: electric streaming and induced potentials; microgravity/tilt (subsurface density changes/surface deformation); seismic wave velocity and attenuation and changes in acoustic impedance (structural boundaries); electrical conductivity/dielectric constants; and magnetic properties. Of all the geophysical methods that can be used in characterization and monitoring, seismic and electrical methods appear to have the most potential, yet have received the least consideration. Both are known to be sensitive to the fluid content and distribution as well as to soil deformation and changes in fluid saturation. Like the hydrologic studies that they are designed to assist, the geophysical methods need considerable development to meet the challenges presented by hazardous waste applications.

To optimize the use of geophysical methods, researchers have identified needs in the areas of instrumentation, processing and interpretation, and computer code development.

New instrumentation is still needed for vadose zone applications, if geophysical methods are to be applied in a cost-effective fashion. Radar can be a very effective high-resolution tool in the vadose zone, especially when applied in a cross-well mode. High-resolution surface seismic measurements need to be made more cost-effective and need to be extended to include recording of shear waves and converted waves so that fluid-content information can be extracted reliably from seismic velocity data.

Many packages are available for processing and interpreting data from individual geophysical field experiments, such as seismic data. Now we need methods for using combined data sets (e.g., seismic and radar together) for deriving the physical properties and lithology throughout the site, and for either directly or indirectly relating the geophysical data to the chemical and microbial properties.

In addition, there is demand for joint inversion schemes utilizing different types of geophysical data (e.g., electrical and electromagnetic data obeying the diffusion and wave equations); and computer codes that can model and handle true 3-D data types (e.g., full-wavefield seismic or electro-magnetic).

Research Priorities: Sensors and Instrumentation	
2004	▪ Understand the effects of emplacement. ▪ Microdrilling and coiled tubing applied as demonstrated minimization of effects at EM sites. ▪ Develop new drilling techniques. ▪ Deploy prototypes in existing boreholes, to decrease cost and improve spatial resolution (goal; 10 sensors, fit in < 2 inch diameter hole). ▪ Characterize the geology and hydrology of a contaminated site at the 10m scale throughout the top 50m of the subsurface using methods that have about a one-month turnaround time for processing and interpretation of field measurements.
2010	▪ Develop minimally invasive methods and begin to correct emplacement effects. ▪ Cheaper smaller robust subsurface location devices. ▪ Deploy field prototype using alternatives to boreholes (CPT, new emplacements, microdrilling, autonomous devices, penetrators, injectable microdevices) (goal: 50 channels, rice grain size hole). ▪ Extend this capability to a resolution of 1m and improve the turnaround time to 1 day, for many field sites.
2025	▪ Develop non-invasive methods, or fully correct for emplacement effects. ▪ Develop injectable sensors and smart sensors. ▪ Off the shelf (goal: continuous spectrum, size of a pore in soil hole, low maintenance, low cost). ▪ Characterize top 50m of the subsurface to 1m resolution in real time for most sites.

2.3 Developing Adequate Models and Simulation Capability

Models are most often thought of as predictive tools for making useful and cost effective estimates of future conditions or, sometimes, reconstructions of past events. Models are used to explore alternative design and operation decisions for characterization, monitoring, and remediation, and to compare and contrast policy options such as remediation and long term stewardship. Subsurface environments are, by nature, incompletely understood. They exhibit a diverse set of important phenomena on a wide range of temporal and spatial scales. They are heterogeneous and difficult to characterize and monitor. For these reasons models also serve a much more fundamental purpose. They are used to synthesize and integrate our understanding of processes, and the coupling between different processes across spatial and temporal scales. Models built for application at a specific site may have more value in this synthesis role than they do in making predictions. The synthesis can be informal and ad hoc or it can be based on formal inversion algorithms and married to both characterization and monitoring data.

Vadose zone predictive and inversion models are based on numerical methods. The resulting computer simulation models only have value if they are pertinent to questions being asked by decision makers, accurately reflect the vadose zone system being modeled, and are obtained in a timely manner. The research identified and discussed in this section is aimed specifically at these numerical models and has been organized into two categories, one related to productivity and the other to predictability.

The usefulness of model results is directly related to the time it takes to build a model, produce a computer simulation, run an inversion, or even display the visualization. The first category of research addresses the question "What new simulation/visualization tools are needed to improve user productivity?" That is, what does the site investigator, the decision-maker, or the researcher need to make better use of their time and resources? These productivity questions are answered by research in three areas: (1) modern software, (2) hardware, and (3) mathematical algorithms all designed specifically for vadose zone applications.

The second category of research addresses the question "What features and capabilities must vadose zone modeling tools possess to both maximize and measure predictability?" Naturally, the answer to this question involves all of the GCBH processes discussed earlier, but, as additions to the productivity questions identified above, there are four areas of modeling research that span all processes: (4) uncertainty, (5) scaling, (6) coupled systems, and (7) integration with characterization and monitoring leading to model validation.

2.3.1 Modern Software: The Vadose Zone Problem Solving Environment

There are a large number of possible conceptual and mathematical models of the vadose zone, and a large and growing number of numerical approximation methods and visualization tools. This sign of maturity aside, modeling of any complex subsurface system is a difficult task, which often leads to compromises in adequacy of the model or numerics used for a particular application. Further, implementing a new model for a complex subsurface system can take months to years of effort, yet is needed routinely. User interfaces are crude and inflexible, several different computer programs must often be inefficiently linked together, and data is only available in a variety of incompatible formats. Such efforts are demanding and time-consuming, and corners are often cut, especially at the research level where most computer software is developed. This situation has led to a large number of limited-use computer programs--i.e., programs used by a small number of people, in many cases only the developer.

These problems can be resolved through the development of a friendly and usable vadose zone problem solving environment (PSE). Formally, a PSE is an integrated software framework that provides the functionality needed for a complex set of tasks using an efficient, high-level interface. Mathematica© and MATLAB© are examples of PSEs, but, for a variety of reasons, they and other commercial products are not suitable for this task. A vadose zone PSE would unify mathematical, scientific, and engineering ideas in a single framework, and allow for rapid set-up of problem specifications and easy manipulation and visualization of data. The development of a PSE for modeling and visualizing subsurface systems, while quite challenging, would significantly enhance the productivity for those developing, improving, or applying models, or relying on them for decisions. It would encourage a greater number of hypotheses to be tested and decision variables to be quantified than is currently possible, inspiring greater confidence in the final decisions. It could and should be integrated with related characterization and monitoring tools.

Research Priorities: Modern Software (The Vadose Zone Problem Solving Environment)	
2004	▪ Develop the basic PSE structure, including a high-level interface; data structure storage and retrieval approach; simulation engine; and graphical back-end; with a common data structure for modeling and characterization/monitoring. ▪ Implement a limited set of state-of-the-art numerical methods. ▪ Restrict to structured grids. ▪ Implement on workstations. ▪ Implement for forward simulations. ▪ Apply on a limited basis.
2010	▪ Build/populate the parameter and location-specific databases. ▪ Mature to a wide range of models, including some multi-scale, coupled models. ▪ Add support for inverse methods and for decision support simulations. ▪ Add support for uncertainty analyses. ▪ Add support for a wide range of numerical methods. ▪ Port to state of the art super computing environments. ▪ Apply on a routine basis.
2025	▪ Add support for the full range of multi-scale, coupled models. ▪ Add support for state-of-the-art numerics, including full spatial and temporal error estimation with automatic grid refinement. ▪ Add support to assess value of information.

	▪ Accept and use as a valuable tool to assist all aspects of the development and application of the science: processes, characterization & monitoring, simulation and prediction.

2.3.2 Modern Hardware: The Environmental Science Computing Initiative

Sheer numerical speed addresses both productivity and predictability. Vadose zone simulations must resolve extreme spatial and temporal gradients, capture multi-scale coupled processes, and handle large data sets. Vadose zone uncertainty analyses, discussed below, dramatically increase the load on computational resources. Today, almost everything is done on fast desktop machines, but limitations on available computer memory and storage, and processor, bus, and network speeds, greatly limits the ability to realistically represent processes, synthesize data, compute forward predictions, or visualize information. Occasionally simulations are run on terraflop machines like the Accelerated Strategic Computing Initiative's Blue and White Machines, but access is limited due to existing priorities and current security precautions. In short, the present generation of hardware available to the vadose zone community is inadequate.

The vadose zone community needs priority access to new state-of-the-art massively parallel computers, like the ASCI's terraflop machines. The vadose zone community would use these machines for research into multiscale processes and process coupling, for characterization, for modeling and prediction, and for visualization. The machines would be housed at one or more national labs thus allowing for priority use by university and national lab researchers working on a wide range of environmental science and technology issues, not just the vadose zone.

There would have to be a dedicated computer support staff to help vadose zone scientists and engineers to prepare their computer programs for parallel processing and port them to the massively parallel computer. This staff could also unite these new computation resources with the proposed vadose zone Problem Solving Environment. The support staff could be patterned after the Center for Applied Scientific Computing staff at LLNL that runs the ASCI Blue and White machines.

Research Priorities: Modern Hardware (The Environmental Science Computing Initiative)	
2004	▪ Build a user community; form a organizing committee ▪ Write the proposal for a multi-teraflop (10^{13} to 10^{14} Flops) Green Machine, to be associated with the Environmental Management Program and/or with the Office of Science. ▪ Complete the design of the Green Machine, including its expected future growth in size and capability. ▪ Purchase and install the Green Machine at a national laboratory; hire staff; begin building (or transferring from ASCI and others) infrastructure like desktop visualization, archival storage, parallel I/O, etc. ▪ Begin training the user community.

2010	▪ Green Machine is used by large variety of vadose zone researchers on studies of processes, characterization/monitoring, and modeling. It is used to study both laboratory and field experiments. It is used in applied studies at DOE environmental management sites. ▪ Extend Problem Solving Environment onto the Green Machine ▪ Form a committee and write a proposal for the next generation of Green Machine. Design the machine, purchase and install.
2025	▪ The next generation Green Machine is in common use for vadose zone problems.

2.3.3 Modern Numerics: Advanced Numerical Algorithms

Although advances in computer hardware will aid the modeling effort, they cannot provide all of the necessary improvements in light of issues like scaling and extreme gradients. Furthermore, the usefulness of model results is not only directly related to the time it takes to produce a simulation and but also to the confidence that a decision-maker can place on the results. The simulation should be accurate, and it should provide some quantified sense as to the likely range and probability of numerical error.

Ongoing basic research on linear and nonlinear solvers, optimization techniques, grid generation, parallelization, visualization, etc. will have a large impact on vadose zone simulation, but must be adapted to its particular needs and resources. Consider two examples. There is a need to tailor linear and nonlinear solution techniques to the vadose zone, including the invention of appropriate preconditioners and multilevel techniques. To capture meaningful space and time scales of important physical and bio-geochemical phenomena, especially non-continuum and chaotic phenomena, new and sophisticated multi-scale numerical algorithms (numerics) are required.

Sources of numerical errors in computer simulations have many origins. Spatial and temporal discretization errors are widely recognized. Refined grids and time steps can reduce these errors. However, in heterogeneous systems like the vadose zone refinement is needed only in certain regions of space and/or time. The computation should automatically and adaptively refine its grid and/or time steps locally as necessary to maintain accuracy, and also unrefine if possible to save computational effort. Time truncation error is particularly difficult to estimate and control. It accumulates from one time step to another, and is often self re-enforcing. However, there is a limit to refinement. As refinement increases, so does rounding error and ill conditioning of the system. Certain types of numerical algorithms contribute other numerical artifacts, such as numerical instability, mass non-conservation, violation of the maximum principle (i.e., creation of nonphysical local maxima or minima in the solution), numerical dispersion, and initiation or non-initiation of physically relevant frontal instabilities. Such errors are often not ameliorated by refined grids and reduced time step intervals, because the mathematical structure itself is improperly approximated. Of particular interest is the ability to resolve features such as extreme spatial and temporal gradients without producing numerical oscillations. These issues must be investigated and resolved within the context of vadose zone applications.

Furthermore, there is a need for algorithms to automatically alert the user to a model's possible numerical limitations, through quantified error range estimates. An even greater development would be algorithms that self-adaptively select a more accurate alternative approach. Numerical error estimates should also be written in terms of measures of interest to decision-makers. For example, estimation of decision variables (mathematically referred to as functionals of the solution) and their variance is more meaningful than merely trying to estimate the norm of the global error. A functional can be overly sensitive to small errors in the solution or data at some point in time and/or space, even though the global error appears to be quite small. Conversely, the functional may be insensitive to such errors, therefore reducing computational effort. Useful algorithms must not only estimate the mean and variance of quantities of interest to a decision maker, but also provide information on the likely error in those quantities generated from a variety of sources, including numerical resolution, inadequate or inappropriate numerics, scaling, and noisy data. These numerical algorithms should be used together with algorithms tracing uncertainty due to the conceptual model, and to uncertain and variable parameters, initial conditions, and boundary conditions.

The algorithms envisioned would be enormously complex and therefore of suspect utility to practitioners and regulators not well versed in numerical techniques and computational issues. They must be ported to the friendly and usable problem solving environment (PSE) discussed above.

Research Priorities: Modern Numerics (Advanced Numerical Algorithms)	
2004	▪ Develop and apply spatial numerical error estimators and adaptive local (spatial) grid refinement algorithms. ▪ Begin estimating numerical error in terms of quantities of interest to decision-makers. ▪ Apply state-of-the art approaches to resolve extreme spatial GCBH gradients, such as fingers and sharp fronts, boundaries, and interfaces. ▪ Adapt, improve and apply state-of-the-art linear and nonlinear solvers.
2010	▪ Develop and apply spatial and temporal error estimators and adaptive local grid and time step refinement algorithms for nonreactive systems. ▪ Complete probabilistic analysis of likely numerical error instead of maximum error. ▪ Using this probability approach begin estimating mean and variance, as well as other numerical error measures, in terms of quantities of interest to decision makers. ▪ Determine criteria for acceptable computational errors based on model uncertainties. ▪ Improve approaches to resolve extreme spatial and temporal gradients, such as fingers and moving sharp fronts, boundaries, and interfaces. ▪ Develop multi-scale mathematical methods and numerics to capture meaningful scales of important GCBH phenomena. ▪ Improve linear and nonlinear solvers to match vadose zone problems and computing environments.
2025	▪ Develop and apply spatial and temporal error estimators and adaptive local grid and time step refinement for reactive systems.

	▪ Estimating maximum, mean, variance, and other numerical error measures in quantities of interest to decision makers becomes standard practice, and is used adaptively to improve simulation results. ▪ Continue to improve approaches to resolve extreme spatial and temporal gradients, such as fingers, and sharp fronts, boundaries, and interfaces. ▪ Develop more sophisticated multi-scale mathematical modeling and numerics, targeted to non-continuum and chaotic phenomena. ▪ Numerical methods are selected automatically, on an appropriate scale, to obtain reduced numerical error in decision variables. ▪ Continue to develop improved linear and nonlinear solvers.

2.3.4 Uncertainty

Uncertainty will always remain an issue with vadose zone models of flow and transport, if for no other reason than the vadose zone is heterogeneous and difficult to characterize and monitor. Uncertainty comes from the incomplete answers to two fundamental questions with which modelers must deal. First, what is an appropriate conceptual model for the intended purpose, one that is not too complex, or one that is not overly simplistic? Second, what are appropriate model parameters (e.g., properties) initial conditions, and boundary conditions? The answers to these questions depend on fundamental research on process and process coupling, and on characterization and monitoring activities. However, it is possible that subsurface systems also have an intrinsic or irreducible uncertainty due to the spatial and temporal stochastic aspect of GCBH processes, and the difficulty of observing these processes without completely dissecting the system. How then is uncertainty traced through the model, and converted into quantities that decision-makers can understand and use?

Uncertainty is now treated primitively, if it is treated at all. Spatial variability of some flow parameters is sometimes simulated by generating simplistic alternative geologic realizations using stochastic methods, or by running Monte Carlo realizations. Recently, these realizations have been conditioned by invasive and non-invasive monitoring and characterization data, which is taking a step in the right direction. Temporal variability due to climate changes can be treated similarly, although historical records are often used instead, and more commonly the flow is considered to be steady. Most conceptual model uncertainty is dealt with informally, by an ad hoc process of deciding whether or not the model is "good enough." While formal inverse approaches to estimate parameters are being developed, there has been much less attention paid to estimating boundary conditions (e.g., climate, meteorology, contamination source), and almost none to estimating appropriate conceptual models. Some conceptual models are useful for answering certain questions but fail miserably in attempting to answer others. An appropriate conceptual model depends not only on the system but the questions being asked--it must be pertinent.

The capacity to estimate and reduce uncertainty will lead to substantial increases in confidence in our ability to make good decisions regarding remediation, waste handling and/or long-term stewardship. While Monte-Carlo simulation is a computationally expensive method, it demonstrates that uncertainty estimation is possible, and conditioning Monte Carlo realizations demonstrates that adding data can reduce uncertainty. Monte Carlo simulation will always be an appropriate tool for some vadose zone applications, but more computationally (and theoretically) efficient alternative sensitivity or uncertainty analyses, possibly based on more sophisticated probabilistic approaches, are needed. There is also need for models to automatically alert the user to possible limitations caused by uncertainty, just as can be done for numerical error. A model could recognize that the conceptualization and data are inconsistent, well beyond the reasonable limits, suggest possible explanations for the inconsistency, and even adapt corrections itself. With formal uncertainty analysis and reduction methods it should be possible to predict the value of new data, to optimize the design and operation of characterization and monitoring activities, and to automatically update a model when new data becomes available. These new developments in uncertainty estimation, reduction, and application should be incorporated into the vadose zone Problem Solving Environment.

Heterogeneity is a major source of uncertainty in vadose zone models. New theories for describing and understanding the spatial and temporal structures of naturally occurring heterogeneities and fluctuations are needed. Existing theories have not yet contributed much to solving basic science or engineering problems. New links between probabilistic description of the media and the probabilistic measures of flow and transport are needed.

Research Priorities: Uncertainty	
2004	▪ Assess the state-of-the-art for methods of uncertainty estimation and reduction, especially for risk analyses. ▪ Prioritize and quantify sources of uncertainty, including choice of conceptual model, geologic heterogeneity; parameter values; and initial and dynamic boundary conditions. ▪ Assess uncertainties contributed by geologic heterogeneity in porous media. ▪ Assess uncertainty contributed by dynamic boundary conditions related to weather and climate variability and predictability. ▪ Assess uncertainties contributed by characterization and monitoring methods, including sample size, sample frequency, etc. ▪ Assess uncertainty contributed by conceptual models of processes, especially overly simplistic or incorrectly scaled process models, and incorrectly coupled processes. ▪ Analyze existing long-term GCBH records to improve conceptual models and reduce their uncertainties and the uncertainties of parameter and boundary condition estimates. ▪ Improve numerical algorithms to increase efficiency of vadose zone Monte Carlo simulations. ▪ Begin testing uncertainty estimation and reduction methods, and their applications, on synthetic test problems, small-scale and meso-scale laboratory experiments, and field scale research sites.
2010	▪ Establish new theories for describing and understanding the spatial and temporal structures of naturally occurring heterogeneities and fluctuations. Use advanced

	geologic modeling to capture both flow-sensitive and chemical spatial heterogeneity. ▪ Evaluate uncertainties for flow models of highly heterogeneous porous and fractured media, affected by the fracture-matrix interactions and, in dry formations, film flow. ▪ Add more realistic treatment of uncertainties caused by surface boundary conditions. ▪ Evaluate effects of uncertainties for invasive and noninvasive field characterization and monitoring methods with different scales and degrees of resolution. ▪ Develop new, more sophisticated and efficient probabilistic approaches to uncertainty estimation and reduction. ▪ Develop methods to display the degree of uncertainty from different sources and in aggregate (processes, coupling, parameters and properties, variability, etc.). Exploit advanced visualization techniques and other sensual (e.g., sound and touch) interactions with models and data. ▪ Develop methods to automatically alert the user to possible limitations caused by uncertainty. ▪ Use uncertainty estimation and reduction methods to predict the value of new data, to optimize the design and operation of characterization and monitoring activities, and to automatically update a model when new data becomes available. ▪ Implement uncertainty methods at several (typical) DOE sites to achieve the goal of using uncertainty estimation and reduction methods as standard practice. ▪ Link the modeling uncertainty analysis with practical problems of a facility designer. ▪ Integrate uncertainty estimation and reduction, with estimation and reduction of numerical errors. ▪ Link to Problem Solving Environment
2025	▪ Incorporate uncertainty in biogeochemical models and models of multiphase flow in fractures and have the models in common use. ▪ Develop uncertainty and inverse methods that self-adaptively suggest modified conceptualizations or parameterizations. ▪ Forward and inverse models accounting for uncertainty are fully integrated ▪ Uncertainty estimation and reduction is standard practice, and use for characterization, monitoring, remediation and stewardship decisions.

2.3.5 Scaling

Mathematical models of processes governing fate and transport in the vadose zone are constructed from mass, momentum, and energy balance statements. These statements depend on constitutive theories that define how flow and transport variables depend on material properties. Examples of flow properties include unsaturated hydraulic conductivity, water retention curves etc., defined for a specific scale. What is not explicitly incorporated are processes at other spatial and temporal scales, and changes in process description as scale changes. While conventional models, such as the Richards equation for flow, are established as useful representations for idealized uniform porous media, no such media occurs in the natural subsurface. Many relevant subsurface properties exhibit multiple nested scales of variability in both space and time. The impact is that the constitutive properties needed for modeling are no longer uniquely defined. Consequently, the constitutive theories relying on property measurements on one scale may be of little or no use to simulation on other scales affected by different flow and reactive transport processes. In this case standard theory becomes inadequate because flow and transport at different scales requires the use of completely different mathematical models. Scaling issues affect boundary conditions as well as the conceptual model and its parameters. Of special concern for vadose zone models is the temporal scaling of highly variable climatic forcing functions, especially given the wide range of interesting time scales for prediction, from a few years for remediation to millennia for stewardship.

The critical questions in scaling include identifying the fundamental scales for different GCBH processes, describing these processes at an observable scale, identifying the relevant or pertinent spatial and temporal scales of concern for decision making, and rescaling the process to fit these parameters. The modeling exercise must also be concerned with how to capture the data needed to answer these questions. Conventional scaling is often based on either physicochemical similitude over different scales, or on contrived assumptions about variability, neither of which are suitable for fate and transport in structured soils and fractured rocks affected by discontinuities and preferential flow phenomena. Research in scaling must address the multiscale nature of not only the processes and the basic properties that control them, but also the disparate scales of observations for characterization, and for fate and transport regulations. The major endpoints envisioned for general capabilities are multidisciplinary scaling of constitutive theories; massive enhancements in deterministic and stochastic scaling tools; hierarchical frameworks for multiple scales of observation/measurement; and comprehensive error analysis and adaptive scaling methods. Some (but not all) focus areas include non-isothermal fate and transport, the occurrence and impact of preferential multiphase flow in the vadose zone, fate and transport in fractured media, and scaling episodically transient reactive transport in multiscale heterogeneous media via generalized coordinate systems. Both general and focused research efforts must incorporate experimentation and measurements on appropriate scales for confirmation of scaling strategies in realistic environments. Finally, the scaling results must be linked to the vadose zone Problem Solving Environment and to advancements in numerical methods.

Research Priorities: Scaling	
2004	▪ Begin developing simulation tools that provide a hierarchy of constitutive theories, simultaneously accommodating multiple processes on different scales. ▪ Complete initial design and development of a Monte Carlo prototype for massive deterministic simulations of scaling issues. ▪ Use scaling to provide quantification of relevant processes by identification of global system variants, such as contaminant trajectories and travel-times, non-aqueous mobile phase geometry's, cumulative reaction histories over multiple time scales, and subsurface ecosystem dynamics. ▪ Develop new deterministic and probabilistic/stochastic approaches to address cross-scale issues that commonly arise in vadose zone reactive flow and transport.

	▪ Begin research into time scaling of reaction systems and of transient, episodic, and boundary value problems.
2010	▪ Refine computational techniques for solving hierarchically-scaled flow and transport models; including testing of these techniques to meso-scale laboratory experiments, small scale field experiments, or other types of measurements of GCBH processes. ▪ Develop scaling methods for flow in generalized (transient and spatially nonuniform) coordinate systems to accommodate processes that vary on multiple spatiotemporal scales. ▪ Develop extensions of the massively deterministic Monte Carlo method to include probabilistic specifications of properties and constitutive hypotheses. Use it as a testbed for evaluation of scaling approaches and for advanced quantification of errors associated with information loss in scaling. ▪ Conduct research to provide a link between constitutive theories and scale of measurement to the quantities of ultimate concern to decision-makers. Link to characterization and monitoring. ▪ Improve scaling algorithms and methods. ▪ Begin to develop links to Problem Solving Environment , uncertainty estimation, and reduction methods.
2025	▪ Broaden the research focus to consider spatiotemporal scaling in support of source-identification, (as well as other inverse problems). ▪ Completely link scaling theory to the Problem Solving Environment, and uncertainty estimation and reduction methods. ▪ Have capability to model strongly transient GCBH processes over multiple time horizons at applied field sites. ▪ Unify aspects of scaling theory.

2.3.6 Strongly Coupled Systems

Strongly coupled systems are those where two or more fundamental GCBH processes must be understood and modeled to accurately represent the flow and transport in the subsurface. Modeling of these coupled non-linear systems is one the most challenging problems in reactive transport. Examples include bioremediation, in situ redox manipulation for removal of Cr, U, and chlorinated hydrocarbons, coupled heat flow and transport for enhanced remediation or radioactively hot waste, and modeling the leakage of highly basic, highly radioactive tank waste solutions at Hanford and other DOE sites. Feedback between non-linear processes can lead to instabilities, like coupled chemical reaction fronts which can become stable or unstable, depending on the nature of the water chemistry and the mineralogy of the permeable material. The current capability to model some of these processes is mostly limited to crude local lumped parameter models which hide much of the complexity, but at the cost of substantial uncertainty. Conventional technology is unable to handle truly coupled processes. Although more advanced models are available to model specific processes, such as adsorption, precipitation, or microbial transformation, the basic scientific understanding of these processes, and especially of their couplings, is currently too weak to make these models much more than useful research tools.

Therefore, developing a basic scientific understanding of the coupled processes themselves is a paramount goal when it comes to modeling this class of problems. Accurate and complete field and laboratory data are also needed to develop, test, and apply the correct models.

Research needed for coupled models can be divided into database needs and constitutive theory needs, as well as computational resources and numerical algorithms that are discussed above. Coupled models require an understanding of the complex interactions among mineral, microbial, and colloidal surfaces and among fluid, solid, and gas phases. Some of these processes include precipitation, surface complexation, sorption, attachment and detachment, multi-domain diffusion, oxidation and reduction (both inorganic and microbially mediated), porosity alteration, and heat flow occurring in a physically and chemically heterogeneous environment. Progress has been made in understanding some individual processes at a basic level, but research prioritization is needed because of the variety of chemicals and reactions involved. Fundamental understanding at a molecular level may allow a few chemicals to be studied intensely and the results expanded to a wider class of chemicals. In parallel, systematic study of coupled reaction systems in bulk phase media can improve understanding of these mechanisms and their manifestation on important coupled processes at the larger scales of observation.

It is important to realize that many problems in reactive transport modeling can be reasonably well addressed by understanding one dominant aspect of the problem. Nonetheless, models need to be available that incorporate all of the relevant processes in order to select the appropriate interactions. Too often the model is selected and processes not included within that model are simply ignored.

Research Priorities: Strongly Coupled Systems	
2004	▪ Identify priority coupled process science needs in vadose-zone fate and transport, review the state of the science in constitutive theory for modeling realistically coupled processes, and initiate research on improved representations of coupled processes of immediate importance. ▪ Update and check reliability of available databases used in coupled models. Make databases universally available with recommendations for usage. ▪ Advance models for coupled systems such as: mechanics of biofilms; multivariate reaction kinetics; saturation and colloid-facilitated transport; and scaling of coupled processes in space and time. ▪ Develop model studies to demonstrate ability of existing coupled models and recommend procedures for usage.
2010	▪ Develop and incorporate improved constitutive theories and parameter databases into coupled models. ▪ Link coupled process models with scaling techniques. ▪ Mechanical-hydraulic and biochemical-hydraulic prototypes available for use. ▪ Test coupled models on synthetic test problems, small-scale and meso-scale laboratory experiments, and field scale research sites. ▪ Implement support for strongly coupled models within the Problem Solving

	Environment.
2025	▪ Improve computational algorithms for coupled processes. ▪ Test strongly coupled models in field applications. ▪ Realistic models are available for all coupled processes important to vadose zone flow and transport, at all important time and space scales. Most models are available as part of the Problem Solving Environment. ▪ Three dimensional fully coupled heat and multiphase flow and transport models are the norm for decision support.

2.3.7 Integration and Validation

The processes and properties that control flow, transport and transformations must be ultimately understood and quantified to form a basis for predictions affecting various decisions. Data from a myriad of characterization techniques, both non-invasive and invasive, will likely be integrated over many iterative cycles to achieve a shared (multidisciplinary) aggregate model whose description consistently honors all data and their uncertainties. Quantities simulated by mathematical models (e.g., pressure, and concentrations) are more readily measured than are many model input parameters (e.g., boundary conditions like infiltration, or properties like hydraulic conductivity and porosity). On the other hand, estimates of model input parameters are often made using inverse modeling, an approach plagued by problems of instabilities, non-sensitivities, and non-uniqueness. Additionally, parameter estimation procedures are still frequently applied without making any probabilistic assumptions. The resulting parameter estimates are then rather meaningless since not much can be said about the relationship between the estimated and true parameter values, and consequently about uncertainties of model predictions. Data collection programs are still regularly designed without studying beforehand parameter sensitivities to measured variables.

Definition models must be integrated with data from characterization and monitoring activities, and validated to ensure that they are performing adequately for the purpose. Data is needed to test the models, and it can come from a variety of sources. Synthetic (computer) test problems provide the only data sets in which all aspects of the system are known. Integrated meso-scale laboratory and field experiments provide a higher level of realism, but with gaps in even the most exhaustive database. Finally, application to site studies is as real as it gets, but leaves even larger data gaps. Models should be tested and validated for each of these kinds of systems, with special attention to comparative studies of various approaches to inverse modeling and estimation.

To improve models, and to assist with integration, new inversion techniques must be developed to estimate parameters, boundary conditions, and the conceptual model. Traditionally parameters (e.g., properties) have received the most attention, usually under steady flow assumptions. Transient data generally reduce the uncertainty of parameter estimates and can overcome identifiability problems. The new methods should couple transient flow and transport processes, together with other characterization information in large scale joint inversions. As part of the of Problem Solving Environment it would also be useful to establish a computational test bed to validate models consistently against relevant data.

Using the synthetic problems, well controlled meso-scale experiments, site data, and possibly other means, models will be tested and validated considering: 1) whether the model consistently honors all available data and understanding of processes; 2) whether the model and data are adequate to make the desired predictions, and 3) where new data would most likely improve model performance. New measures of model performance will be needed.

To apply models to sites, the entire modeling approach should be integrated with data through geographical information systems (GIS). GIS provides tools and methods for managing data in space and time including special interrelationships and uncertainties. GIS tools could be an integral part of the Problem Solving Environment (PSE), or simply linked to it. Site considerations also suggest the design, building, testing, and updating of a Shared Vadose Zone Model, that contains a subset of features of the PSE sufficient for dealing with most vadose zone problems. It, rather than the parent PSE, could become the main vehicle for model application and decision support at sites.

Research Priorities: Integration and Validation	
2004	▪ Develop sampling event models and instrument response to improve instrument design for invasive and non-invasive characterization and monitoring methods. ▪ Develop new parameter estimation inversion techniques for transient flow and transport processes. ▪ Develop new model testing and validation tools. ▪ Define synthetic test problems and conduct comparative studies of inverse modeling and model validation for these problems.
2010	▪ Complete integrated meso-scale laboratory and field experiments to provide databases for specific model testing and validation. ▪ Establish a computational test bed as part of the Problem Solving Environment (PSE) to assist model testing and validation. ▪ Improve and apply inverse modeling and probabilistic uncertainty estimation methods to the field and meso-scale laboratory experiments. ▪ Link GIS (geographical information systems) to the PSE. ▪ Couple uncertainty analysis to management models and PSE to help make decisions concerning characterization, monitoring, and remediation design and operation. ▪ Begin applying model testing and validation methods approaches to DOE sites.
2025	▪ Establish better inverse methods for parameters, and boundary conditions and models, and their uncertainties. Use as a guide for characterization and monitoring. Test using synthetic problems, experiments, and site data. ▪ Develop and apply a highly integrated Shared Vadose Model, a subset of the PSE, where the value of new data is continually assessed as part of an observation–prediction–decision support (cost analysis) process. ▪ Models are finally a "bridge" to communicate findings between scientists and policy-makers.

3.0 CONCLUSIONS AND RECOMMENDATIONS: SETTING PRIORITIES, INTEGRATING APPROACHES, AND BUILDING INFRASTRUCTURE

3.1 Setting Priorities and Sequencing Research

Not surprisingly, there is much overlap in the broad areas of research identified as priorities by the Roadmapping Workgroups. This overlap can be captured by organizing the Workgroups' interests into four categories—the research required to: (1) understand the geological, chemical, biological and hydrological framework; (2) understand fluid flow and transport; (3) integrate data, understanding and models within an experiment, and generalize the results beyond a single experiment or site; and (4) develop new tools and techniques. These categories, and their connection to the research priorities discussed by the workgroups, are illustrated in Figure 3.

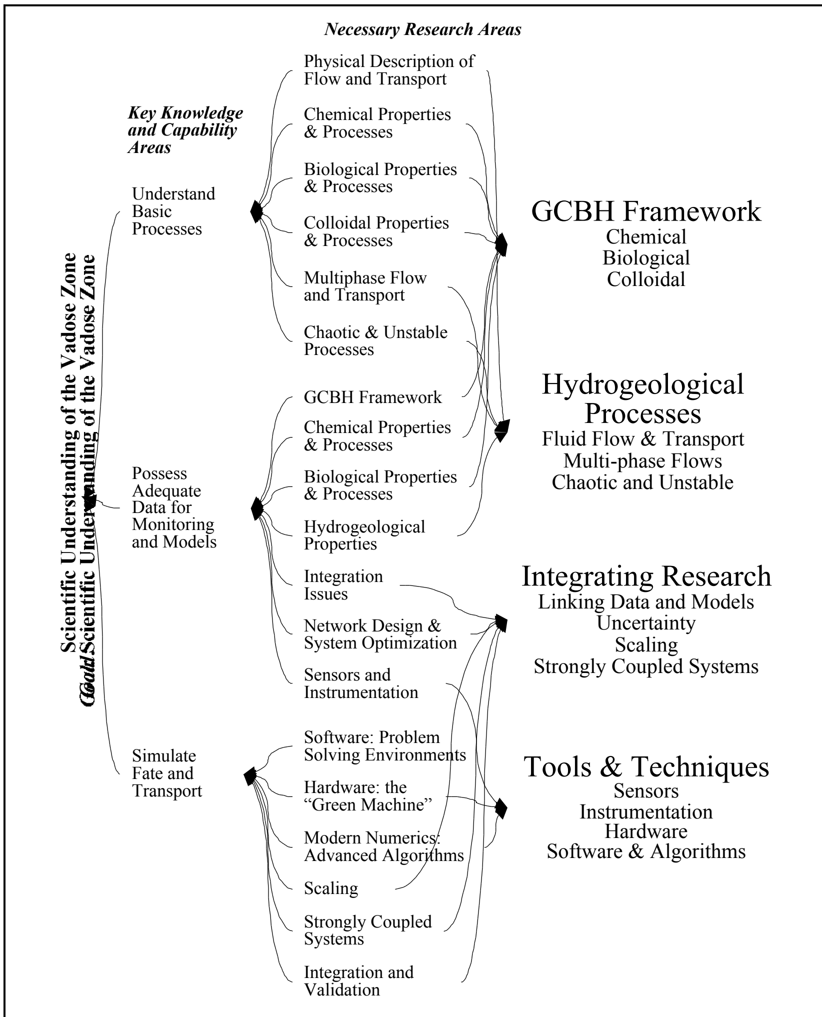

Figure 3.

The research priorities for each category for the near-term (i.e., for 2004), the mid-term (2010) and the long-term (2025) are detailed in Appendix C, and represent a rough sequencing of the activities necessary to achieve this roadmaps goals. Taken together, these research priorities represent the guideposts by which the vadose zone S&T community can organize themselves to meet DOE's pressing need for an adequate and reliable scientific understanding of the vadose zone and the tools and techniques that will allow for a high degree of certainty in predictions. Additional work to focus and sequence the research priorities identified in the Roadmap will continue within the vadose zone science and technology community in FY01. These will include workshops dedicated to reaching this third level of understanding, and national meetings and technical reviews aimed at soliciting feedback from the affected stakeholders.

3.2 Integration within Experiments and Generalization to Other Sites

Integration and integrating research is critical if the vadose zone research community is to move forward in its ability to understand the basic processes at work in the vadose zone, possess the data necessary for monitoring contaminant migration and building computer simulations, and adequately model and predict contaminant behavior and fluid flows. These, when combined, will provide the scientific basis for decision-making with respect to the vadose zone. To date, this integration has been hampered by 1) difficulties correlating our understanding of basic vadose zone processes with characterization and monitoring data, and with computer models of contaminant fate and transport within *individual* sites or experiments, and 2) difficulties integrating information such that knowledge garnered at one laboratory or site can be generalized to other sites and experiments.

The solutions to these key integration barriers lie both in what should be addressed (technical priorities) and in how vadose zone research should be conducted (the structure and focus of the research plans and programs developed by DOE's program offices).

Integration within Experiments. There is great consensus that the ability to integrate measurements and interpretation from different disciplines such as biology, geochemistry, hydrogeology, geology, geophysics, and geomechanics is fundamental to our ability to provide a solid scientific understanding of the vadose zone. An integrated understanding of these disciplines will enable the selection of characterization and monitoring methods appropriate to a specific site with optimized sensor location and frequency of measurement. It will also lead to the collection of data at levels of precision that makes sense from a cost-to-benefit standpoint for each of the relevant parameters, and for the development of models that more accurately account for all the processes and properties that are important.

Increasing the interplay between modeling and data gathering in experimentation is another crucial element of integration in vadose zone research. Just as characterization and monitoring strategies may be improved with guidance from newer and more relevant computer models of vadose zone behaviors, the ability to discriminate among alternative conceptual models and to improve on them will depend on the validation of their predictive capabilities against characterization and monitoring data.

Scaling. The ability to extrapolate results from laboratory experiments to field experiments or vice versa is one of the most pressing challenges facing vadose zone research. Solving scaling problems will be central to this ability. Scaling issues come in a number of forms: first, the large spatial variability of hydraulic properties in natural geologic systems over a wide range of scales renders estimation of hydraulic parameters and their spatial correlation difficult. Second, the instrument and network scales used to characterize and monitor sites vary among, and even within, geophysical methods and thus create questions about how measurement-related scales affect obtained values, and how to appropriately incorporate or combine different types of measurements.

The addition of computer models to this picture makes scaling issues even more thorny: because many relevant subsurface properties exhibit multiple nested scales of variability in both space and time, simulations based on property measurements on one scale may be of little or no use to simulations on other scales affected by different flow and reactive transport processes. In this case standard theory becomes inadequate, because flow and transport at different scales requires the use of completely different mathematical models.

Advances in our ability to understand how to scale measurements collected in the laboratory for use in the field, how to utilize petrophysical relationships developed in the lab with field data, and how to integrate different data that sample different volumes within our computational and predictive models are key to dramatically improving our scientific understanding of the vadose zone.

Uncertainty. Uncertainties in the quantitative understanding of flow and transport of complex contaminant mixtures at DOE sites have many sources in data collection methodologies, modeling techniques, and gaps in the fundamental knowledge of basic properties and processes. Often, the relative contributions to uncertainty arising from these various sources are unknown. Refinement of fundamental knowledge, data collection techniques, and modeling approaches to reduce uncertainty is expensive, and the more certain we wish our monitoring and predictive capabilities to be, the more expensive the refinements become. These costs are counterbalanced by those arising from over design of engineering facilities needed to compensate for uncertainty, and by those arising from public mistrust when uncertainty is inadequately recognized and predictions fail. Thus, two challenges face the vadose zone research community: understand and reduce sources of uncertainty; and decide how much certainty is necessary to minimize over design and to avoid unpleasant surprises. For the first challenge, a next generation of characterization, monitoring and modeling capabilities is needed that provide better quantification of estimate uncertainty due to measurement errors, inversion errors, and use of estimated petrophysical relationships. We must understand how use of multiple co-located data sets reduce ambiguity and uncertainty associated with parameter estimates. Existing stochastic methodologies handle some issues involving uncertainty, but further refinement, testing, and extension of these approaches is needed, with particular regard to issues of scale. The second challenge, determining the level of scientific certainty required to make good decisions regarding the vadose zone, is not the purview of the research community alone-but will require substantial dialogue with the larger DOE, policy, and stakeholder communities.

Integrated Field Experiments. The wide-ranging integration needs discussed above have led the roadmapping team to consensus in suggesting that DOE should consider not only what vadose zone research to sponsor, but how that research should be conducted. Thus, the team believes that a combination of multidisciplinary, laboratory, numerical, and field scale studies will be necessary to test specific hypotheses and relate all aspects of vadose zone behavior. Specifically, the team suggests that the use of integrated field experiments, organized to address vadose zone questions that are most important to DOE's ability to assess and predict contaminant fate and transport, will be the key to fostering the integration of research approaches and knowledge necessary for revolutionary advances in the application of vadose zone research.

These integrated field experiments would enable researchers of many disciplines (e.g., chemistry, biology, mathematics), with many goals (e.g., sensor, instrument or model development), and working at many scales (e.g., laboratory, bench, and field) to tackle different portions of one overarching problem. For example, understanding the transport of radioactive contaminants from disposal pits in an arid environment, or understanding fluid flows and transport in highly fractured media. Their results would be far more easily interpreted in the context of other research, and would begin to build a body of experimental results that bridge scale, application, and discipline, and are more generalizable to other similar vadose zones.

3.3 Necessary Infrastructure

In addition to providing a roadmap of research priorities spanning the next quarter century, and recognizing the creation of integrated field experiments as key to advancing vadose zone research, the roadmapping team believes that additional infrastructure will be needed.

3.3.1 Sources of Useful Information: Data and Model Libraries

There is much consensus within the vadose zone research community on the need for a broader and more effective means for disseminating the information that currently exists. As one participant put it, "our first steps in addressing vadose zone issues should be to apply the knowledge we have now." To date, widespread distribution and application of this information has been greatly hampered by 1) the lack of a convenient means for sharing vadose zone experimental data and results, and 2) differing standard practices in data collection from site to site, and experiment to experiment.

Currently, each DOE facility maintains it's own standard operating procedures (SOP's), and the majority of these facilities do not share their data with other DOE facilities. A case in point: for many of the case studies discussed in *The Vadose Zone: Science and Technology Solutions*[8], its publication represented the first widespread dissemination of the numerous experiments and results from DOE facilities. This practice of maintaining facility-specific SOP's results in substantial additional expense and very little opportunity for developing a common scientific framework for understanding vadose zone parameters and processes. Thus, many within the vadose zone research community have called for a "data library" (whether virtual or real) that would place the results of current experiments in context and in the hands of those who might benefit from them.

Similarly, many roadmap participants have recognized the lack of, and need for, access to unifying models that can be used for the vadose zone. To date, the flow and transport simulation capabilities that have been developed are very site-specific. The notion of a reliable and commonly used solute transport model which would allow supporting models to converge into a solution does not exist at this time, but is desirable in the long-term.

[8] formal citation to come

3.3.2 Computing Power: "The Green Machine"

Both the vadose zone data visualization capabilities and the predictive simulations envisioned in this roadmap will require computing power on a scale similar to that of the massively parallel computers used for DOE's Accelerated Strategic Computing Initiative (ASCI). ASCI is designed to provide the ability to assess, evaluate, maintain, and prototype nuclear weapons and weapons components through computer simulations and without actual nuclear testing, which is now banned. The need for these simulations has always been obvious—researchers can never know exactly what happens during a nuclear detonation merely by exploding nuclear weapons, yet fundamental understanding of the micro-scale and macro-scale reactions and processes are crucial. DOE's computational codes are now capable of simulating extreme and extremely complex events (nuclear fusion among them) in micro-second timeframes, with considerable accuracy.

Modeling vadose zone behaviors is a problem of similar difficulty. Thus this roadmap identifies the need for an environmental science computing initiative, centered around a state-of-the-art massively parallel machine, a "Green Machine," that would be available for priority use by university and national laboratory researchers working on the problems of understanding and predicting subsurface contaminant fate and transport.

3.3.3 National Leadership: A Vadose Zone "Czar"

The desire for a strong scientific basis for making public policy, regulatory, remediation or long-term stewardship decisions related to the vadose zone is not limited to DOE. The number of stakeholders is large. For example, other federal agencies, notably the U.S. Departments of Agriculture and Defense, the United States Geological Survey and the Environmental Protection Agency are major players—sometimes as problem holders, sometimes as policy makers, sometimes as research sponsors, and sometimes as all three. State, local and tribal governments also have a stake in the decisions made regarding the vadose zone. As do individual site managers, whose remediation, waste handling and long-term stewardship decisions are governed by a web of regulatory, policy, budget and scheduling constraints and are creating a quilt of unique and inconsistent standard operating procedures across the country.

The research community is no less varied: it encompasses a wide range of academic disciplines in settings that span the public, university and private sector. Here, the interplay between regulations as drivers of technology research and development, and technology as a driver of regulation adds enormous complexity to the challenge of moving the entire field of vadose zone research forward. Public perception of risk and its acceptable levels adds unpredictability to this mix. Finally, the nature of vadose zone processes themselves: complex, incorporating large volumes of media, and undergoing transformations that may occur very rapidly or in timeframes than span centuries, do not lend themselves to quick, simple solutions.

All these factors point to the need to address vadose zone issues as the national concern that they are. First and foremost, leadership will be required to create and guide a multi-agency, multi-year, long-term and integrated approach to the vadose zone challenge. Next, many policies will require change. For example, federal and state regulatory systems that currently overlook the vadose zone entirely must evolve to provide incentives to building “early warning” monitoring and prediction capabilities; and current DOE funding cycles which are conducted on an annual basis must be revised to be more compatible with research on contaminant migration, which often occurs only over decades. Finally, some consistency in approach across agencies and among researchers must be devised and adopted. This might take the form of consensus standards, similar to those developed by the American Society for Testing and Materials and approved by its 33,000 members. Such an approach is in keeping with current Technologies Transfer Act Amendments of 1996, signed by President Clinton, that state that federal agencies will use “consensus standards” in carrying out their fundamental responsibilities.

3.3.4 Final Thoughts

After reviewing DOE’s stewardship program the NRC concluded, “a long-term commitment to both basic and applied research is needed. This research must address...basic scientific questions about the behavior of wastes in the diverse environments of the nations nuclear waste sites.” The authors of that report recognized that “stewardship and science are interdependent a must be managed together. Site stewardship that includes the monitoring and encouragement of emerging new technologies and scientific breakthroughs for their relevance to further reducing the risks associated with residual contaminants would, over the long run, decrease the potential consequences of stewardship failures. To this end, the NRC recommended, among other activities, that DOE “undertake necessary scientific, technical, and social research and development.” This roadmap provides the first steps along the path suggested by the NRC.

APPENDIX A: ROADMAP CONTRIBUTERS

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